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DECISION
of 8 March 1996

Case Number:	T 0288/94 - 3.4.2
Application Number:	87110763.7
Publication Number:	0256359
IPC:	B01D 53/36, B01J 23/22, B01J 23/24, B01J 35/10
Language of the proceedings:	EN

Title of invention:
Catalysts and use thereof for removing nitrogen oxides in exhaust gas

Patentee:
MITSUBISHI PETROCHEMICAL CO., LTD., et al

Opponent:
BASF Aktiengesellschaft, Ludwigshafen

Headword:
-

Relevant legal provisions:
EPC Art. 56

Keyword:
"Inventive step (yes)"

Decisions cited:
-

Catchword:
-

Case Number: T 0288/94 - 3.4.2

DECISION
of the Technical Board of Appeal 3.4.2
of 8 March 1996

Appellant: (Proprietor of the patent)	MITSUBISHI PETROCHEMICAL CO., LTD. 5-2, 2-chome, Marunouchi Chiyoda-ku Tokyo 100 (JP)
Representative:	Hansen, Berd, Dr.rer.nat. Hoffmann, Eitle & Partner Patentanwälte Postfach 81 04 20 D-81904 München (DE)
Respondent: (Opponent)	BASF Aktiengesellschaft, Ludwigshafen -Patenabteilung - C6- Carl-Bosch-Strasse 38 D-67056 Ludwigshafen (DE)
Representative:	Dost, Wolfgang, Dr.rer.nat., Dipl.-Chem. Patent- und Rechtsanwälte Bardehle . Pagenberg . Dost . Altenburg . Frohwitter . Geissler & Partner Postfach 86 06 20 D-81633 München (DE)
Decision under appeal:	**Decision of the Opposition Division of the European Patent Office posted 14 February 1994 revoking European patent No. 0 256 359 pursuant to Article 102(1) EPC.**

Composition of the Board:

Chairman: E. Turrini
Members: R. Zottmann
L. C. Mancini

Summary of Facts and Submissions

I. The Appellant (Patentee) lodged an appeal against the decision of the Opposition Division revoking patent No. 0 256 359 (application No. 87 110 763.7).

The Opposition Division held that the subject-matter of the independent claim of the main and the auxiliary request did not involve an inventive step with respect to the prior art disclosed in document

D1: DE-A-3 433 197.

II. In a reply to the grounds of appeal the Respondent (Opponent) mentioned further prior art documents:

D4: Chem.-Ing.-Techn. 56 (1984), 6, pages 455 to 463,

D5: Chem.-Ing.-Techn. 50 (1978), 8, page 637,

D7: EP-A-0 073 703 and

D8: H.-G. Franck, A. Knop: "Kohleveredlung: Chemie u. Technologie", Springer-Verlag, Berlin, Heidelberg, 1979, pages 21 to 23

and requested that the appeal be dismissed.

III. The Appellant requested that the decision under appeal be set aside and the patent be maintained with the documents according to a main request or an auxiliary request. The documents according to the main request are the patent documents.

The Appellant requested oral proceedings unless the claims of the main request are accepted.

IV. Claims 1 and 10 of the main request (identical with claims 1 and 10 as granted) read as follows:

"1. A catalyst having a total pore volume of 0.1 to 0.7 ml/g of the catalyst and containing a first group of many pores having a diameter of 1 x 10 nm to less than 1 x 102 nm and a second group of many pores having a diameter of 1 x 102 nm to 1,2 x 104 nm, the pore volume of the first group being at least 10% based on the total pore volume of the first group and the second group, said catalyst comprising titanium and a base metal other than titanium as metal elements of catalytically active ingredient, and the atomic ratio of titanium to the base metal being 1:0.001-1."

"10. Use of the catalyst according to any of the preceding claims for removing nitrogen oxides in an exhaust gas."

Claims 2 to 9 are dependent on claim 1.

V. The arguments of the Appellant can be summarized as follows:

It is conceded that the invention differs from D1 mainly in the pore volume. This difference is, however, both substantial and characteristic. Moreover, the pore volume deduced from Figure 1 of D1 has so far been calculated erroneously by adding the pore volumes corresponding to the measuring points indicated on the curve. The value of 2.8 ml/g obtained in this way therefore merely reflects the volume contributed by the pores with diameters corresponding to the measuring points. The contribution made by pores with a pore diameter between two measuring points was not taken into account in said calculation. The error introduced by this calculation becomes even more significant when considering that the scale on the abscissa is logarithmic. In a more refined analysis (see ANNEX 1 of the letter dated 21 June 1995) it was found that Figure 1 relates to a catalyst with a total pore volume of at least 5.4 ml/g which is thus approximately eight times the pore volume according to the upper limit of the pore volume range according to claim 1.

Figure 1 represents the only embodiment from which the total pore volume can be derived and said pore characteristic cannot be allocated to any of the examples given in D1.

High porosity aimed at in D1 is an essential feature of the teaching of D1 and a skilled person would have no incentive to lower this porosity and certainly not to turn to a porosity eight times lower. D1 clearly teaches increasing the porosity in order to ease the flow of gas, but it pays no attention to the life-span of the catalyst nor to the problems associated with blocking and poisoning.

From an appropriate experiment on the basis of Example 1 of D1 where it was tried to extrude the catalytical material instead of using paper-making and corrugation steps to form the catalyst as in Example 1, it is evident that the intended extrudate cannot be produced (see ANNEX 2 of the letter dated 21 June 1995). Thus the skilled person cannot arrive at the present invention merely by extruding the base material.

D8 makes no mention of As being present in the exhaust gases or in the ashes, does not attend to the poisoning or blocking of the catalyst and thus the combination with D1 is not obvious.

VI. The arguments of the Respondent (contained in his single letter dated 8 November 1994) can be summarized as follows:

The bimodal pore distribution of Figure 1 of D1 is very similar to that of the patent in suit. Figure 1 describes an example of the structure of the catalyst according to the invention and can thus be seen as being typical. The catalyst of the patent in suit differs from the embodiment of Figure 1 of D1 only in the total pore volume which is ca. 2.8 ml/g, whereas the pore volume of the attacked patent is in the order of 0.35 ml/g.

D1 discloses the treatment of flue gases - e.g. of combustion devices - containing fly ashes. The most important fuel which produces exhaust gases containing fly ash is coal. Such fly ashes generally contain a considerable amount of Ca and a less amount of As. Reference is made to D8. Furthermore, the poisoning activity of As with respect to catalysts and the advantages of a bimodal pore distribution with respect to said poisoning are well known in the art. It is, therefore, the aim of D1 to treat also gases containing fly ash and thus also Ca and As. Due to the bimodal porosity distribution the skilled person is well aware that the catalyst has advantages with respect to poisoning, for example by As; reference is made to D5 and D7. D4 teaches that the ratio of small and large pores can be adjusted by the addition of organic fibres.

The pressure loss of a catalyst - the lowering of which is an essential aim of D1 - is only a function of its geometrical form but not of its porosity. Therefore, to improve the stability of the catalyst of D1, the skilled person would consider a lowering of the pore volume. When starting from the catalyst according to D1 and extruding the catalytic composition using high pressure and adjusting the bimodal pore distribution according to D1 by adding organic fibres, the considerably reduced porosity and thus a catalyst according to the attacked patent would be obtained.

Reasons for the Decision

1. The appeal is admissible.

2. *Novelty of claim 1 of the main request*

D1 describes a catalyst for the reduction of nitrogen oxides (NOx) in an exhaust gas such as from boilers etc. The catalyst comprises Ti and a base metal other than Ti (see claim 1: Cu, Fe, V, W and/or Mo). The atomic ratio of Ti and said base metal comes within the range of claim 1 (see all Examples 1 to 5). Though Ti is used as a carrier, it must be regarded as having the same catalytic activity since it is - as in the patent in suit - preferably precipitated from metatitanic acid (see in D1 particularly the paragraph bridging pages 15 and 16). The catalyst base structure is obtained by a process like a paper-making process and by corrugation.

Figure 1 (described on page 8 lines 29 to 31 and page 20 lines 22 to 30) represents the only embodiment of D1 from which the pore diameter distribution and the total pore volume of the (finished) catalyst - this quantity always means: total pare volume per mass of the finished catalyst - can be taken. The pore volumes of claim 1 and of Examples 1 to 5 refer only to the unfinished catalyst. The pore characteristics of Figure 1 cannot be allocated to any of said Examples as no such reference can be found throughout the description and the characteristics of manufacturing method of the catalyst according to the embodiment of Figure 1 are unknown and thus cannot be compared with those of said Examples.

The pore diameter distribution of Figure 1 comes within the ranges as defined by claim 1 of the attacked patent (see Figure 1 of D1 and Figure IV of ANNEX 1 of the letter of the Appellant dated 21 June 1995). However, the total pore volume is considerably higher. Even the erroneously calculated total pore volume of 2.8 ml/g used for the considerations up to the Appellant's refined calculations (see section V above) is four times the total pore volume of the upper limit of claim 1 (0.7 ml/g). Said refined calculation, which has not been contested by the Respondent and which is in the opinion of the Board of Appeal correct, results in a total pore volume of at least 5.4 ml/g. It is thus almost eight times said upper limit.

Therefore and since D8 and the other documents cited during the opposition procedure are much less relevant than D1, the Board has reached the conclusion that the subject-matter of claim 1 is novel.

3. *Inventive step of claim 1 of the main request*

3.1 It follows from section 2. above and is undisputed that the nearest prior art with respect to the subject-matter of claim 1 is the embodiment of Figure 1 of D1 and that said prior art is distinguished from the subject-matter of claim 1 by the total pore volume.

3.2 In spite of the considerably lower pore volume of 0.1 to 0.7 according to claim 1 (and even only up to 0.45 ml/g of the examples) of the attacked patent with respect to the nearest prior art - which is, apparently, predominantly a result of the kind of manufacturing: by extrusion -, the obtainable effectivity of the NOx conversion of up to 98.3% (see Table 3) seems to be very satisfying. This is also the case when said effectivity is compared with the available results of the catalysts of D1 (see there "TABELLE I").

D1 is silent about the resistance of the catalyst against poisoning and/or blocking by As and/or Ca compounds. It is known to the skilled person that the more large pores (above ca. 100 nm) are present the higher said resistance will be. As according to the nearest prior art the pore volume having said large pores is about 2 ml/g (see Figure IV of the said ANNEX 1) and thus considerably higher than that of claim 1 (at the utmost 0.64 ml/g), it can be assumed that said resistance is at least as high as that of the catalyst according to claim 1 of the patent in suit.

The mechanical strength of the catalysts both of D1 and the patent in suit is assessed by measuring the abrasion. The measuring methods of D1 and the patent in suit are, however, so different that the results cannot be compared. Nevertheless, the skilled person will get from D1 the impression (see "TABELLE I", page 8 second paragraph, page 22 second paragraph, pages 33 and 35) that the abrasion of the examples according to the invention has reached a satisfying level (ca. 0.2 g) as compared with the comparative examples (0.34 to 4.85 g). In the description of the attacked patent - see page 5, lines 20 to 31 - it is stated that, if the portion of the total pore volume with said large pores is very high, the abrasion resistance of the catalyst is decreased. However, the results of the abrasion test of D1 do not support such a relationship for catalysts of the type as described in D1: The manufacturing methods of the catalyst base structure of Examples 1 (where the starting material does not comprise organic fibres), 2 and 4 (the starting material of both Examples 2 and 4 comprise organic fibers to be burnt during the calcination step) are quite similar - of course except for the addition of organic fibres. The large pores originate from the catalyst base structure (inorganic fibres and burnt organic fibres if present in the starting material), the small pores from the coatings. Therefore, the catalyst base structures of Examples 2 and 4 have higher pore volumes (calculated from page 24, line 35 to page 25, line 4 as 2.8 ml/g and from 27 lines 20 to 25 as 3.42 ml/g) than the base structure of Example 1 (calculated from page 23, lines 19 to 30 as 1.46 ml/g), presumably due the addition of inorganic fibres. Nevertheless, the abrasion of the catalysts of Examples 2 and 4 ("TABELLE I: 0.18 and 0.19 g) is lower than that of Example 1. Therefore, it is at least quite unsure whether a reduction of the pore volume of the catalyst of D1 would lead to less abrasion and thus to higher mechanical strength.

It seems, therefore, that the only advantage which can unambiguously be derived by the skilled person when comparing the nearest prior art and the subject-matter of claim 1 consists in the following: The combination of pore parameters of claim 1 makes it possible that other, less laborious manufacturing methods entailing a lower pore volume, e.g. the extrusion of a suitable mixture of all components of the catalyst, can be taken into consideration without jeopardizing the satisfying catalyst properties.

3.3 The problem underlying the solution according to claim 1 is therefore to further develop the catalyst of the nearest prior art in such a manner that a less laborious method can be used to manufacture the catalyst without undue worsening of reactivity and lifetime.

3.4 To solve said problem, the skilled person would not envisage reducing of the pore volume of the catalyst, let alone to such an extent as necessary to arrive at a catalyst according to claim 1. Such a reduced pore volume of about one eighth would e.g. probably be obtained if the skilled person tried to manufacture a catalyst by extrusion as in the patent in suit. It is stated in D1 (see page 4 last paragraph to page 5 second paragraph) that such a catalyst has disadvantages, above all a reduced catalytic activity due to a lower porosity. Moreover, the experiments of the Appellant (see section V above) show that at least a catalyst as in Example 1 of D1 cannot be produced by extrusion moulding. A porosity as high as possible is a quite essential concern of D1 and is, according to D1, a prerequisite for high catalytical activity (see for example the problem underlying the invention of D1 on page 8 second paragraph and page 4/5, page 10/11, page 12, lines 14 to 20, page 18 second paragraph, page 20, paragraphs 2 and 3 and page 22, lines 19 to 25).

Even if tests showed that the mechanical strength of the catalyst of the nearest prior art is worse than that of the attacked patent and need improvement, the skilled person would, in view of the fact that D1 stresses the importance of high porosity, not envisage reducing the pore volume by a factor of eight. He would rather try to obtain mechanical strength by other measures, e.g. by a suitable choice of the carrier and/or of binding agents and/or he would try to reduce abrasion by a change of characteristics of the exhaust gas flow and/or a removal of the solid components of the exhaust gas before they reach the catalyst or he would tolerate a moderate mechanical strength of the catalyst.

3.5 The Board has also considered the remaining documents on file and found them non-prejudicial to claim 1 alone or in combination with D1. Therefore, said claim involves an inventive step within the meaning of Article 56 EPC and consequently said claim does not infringe Article 52(1) EPC.

4. Since the independent use claim 10 of the main request, due to its direct or indirect reference to claim 1, contains all features of claim 1, a corresponding reasoning leading to the same result is valid for claim 10, too.

5. The dependent claims 2 to 9 of the main request concern particular embodiments of the subject-matter of claim 1 of the main request. Therefore, they do not infringe Article 52(1) EPC, too.

6. Considering that the main request is allowable, the auxiliary request and the conditional request of the Appellant for oral proceedings need not be discussed.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The patent is maintained unamended.

The Registrar: The Chairman:

P. Martorana E. Turrini