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D E C I S I O N
of 16 September 1999

Case Number: T 0720/96 - 3.3.5

Application Number: 88312315.0

Publication Number: 0323195

IPC: B01D 53/36

Language of the proceedings: EN

Title of invention:

A process for removing nitrogen oxides

Patentee:

Babcock-Hitachi Kabushiki Kaisha

Opponent:

Siemens AG Abtlg. ZFE GR PA 3
Degussa-Hüls Aktiengesellschaft Patente und Marken Standort
Marl

Headword:

Removing nitrogen oxides/BABCOCK-HITACHI

Relevant legal provisions:

EPC Art. 54(1), 56

Keyword:

"Novelty - yes, selection from two independent groups"
"Inventive step - yes, non-obvious solution of a known
technical problem"

Decisions cited:

T 0279/89, T 0012/81, T 0164/92, T 0666/89, T 0181/82

Catchword:

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Boards of Appeal

Chambres de recours

Case Number: T 0720/96 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 16 September 1999

Appellant (01):
(Opponent)

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Appellant (02):
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Respondent:
(Proprietor of the patent)

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Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 24 June 1996
rejecting the oppositions filed against European
patent No. 0 323 195 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: R. K. Spangenberg

Members: G. J. Wassenaar

J. H. van Moer

Summary of Facts and Submissions

I. The appeal is from the decision of the Opposition Division to reject the oppositions and maintain European patent No. 0 323 195 with Claims 1 and 2 as granted. Claim 1 of the patent in suit reads as follows:

"A process for removing nitrogen oxides from exhaust gases containing nitrogen oxides and volatile metal compounds which process comprises contacting the exhaust gases in the presence of ammonia with a catalyst comprising titanium oxide characterised in that the catalyst further comprises a composite oxide of vanadium and at least one of molybdenum and manganese, said composite oxide of molybdenum and vanadium is expressed by the formula MoV_xO_y wherein x represents 0.5 to 3 and y represents 3 to 10, and said composite oxide of vanadium and manganese is expressed by the formula $\text{Mn}_a\text{V}_b\text{O}_c$ wherein a represents 1 to 4, b represents 1 or 2 and c represents 3 to 9."

II. In the contested decision the following prior art documents were mentioned:

D1: US-A-4 071 601

D2: DE-C-2 458 888

D3: Surface Technology, 9 (1979), 195-202

D4: DE-B-1 253 685

EH2: EP-A-0 220 416.

III. In their statements of the grounds of the appeal, the appellants maintained that the product according to granted claim 1 lacked novelty and inventive step. In their argumentation during the written and oral proceedings they further relied on the following newly filed documents:

D5: VGB Kraftswerktechnik, 65 (8), 1985, pages 753-763

D5a: Kirk-Othmer, Encyclopedia of Chemical Technology, Third Edition, Volume 6, pages 224, 228, 247-248, 273-275

D6: Römp Chemie Lexikon, Eight Edition (1979), Volume 1, pages 511-512

D7: Ullmann's Encyclopädie der technischen Chemie, Fourth Edition, Volume 12 (1976), page 570

D8: Römp Chemie Lexikon, Ninth Edition (1990), pages 2267-2277

D9: Römp Chemie Lexikon, Ninth Edition (1989), pages 251-253

D10: US-A-4 377 118.

Further new evidence was submitted in the form of a report of a reworking of Example XIV-1 of D2.

IV. Oral proceedings took place on 16 September 1999. The novelty objection was based on D1 and D2. The appellants argued essentially as follows:

Both D1 and D2 implicitly disclosed the treatment of a gas comprising volatile metal compounds. In this respect reference was made to D5a, D6, D7, D8 and D9. The catalyst used in the process of claim 1 was taught by D1. The catalyst used in Example XIV of D2 was essentially the same as that used in the examples of the patent in suit.

With respect to inventive step both appellants agreed that D1 represented the closest prior art. They essentially argued that even if there was any difference there was no proof that the claimed process solved any technical problem. The comparative examples did not show any advantage over the prior art and the catalysts prepared according to the examples in the patent in suit did not necessarily have the structure as required by claim 1. Reference was made to the following decisions of the Boards of Appeal: T 279/89, T 12/81 and T 164/92.

- V. The respondent maintained that the subject matter of the granted claims was new and involved an inventive step over the available prior art. Neither D1 nor D2 disclosed the treatment of waste gases comprising substantial amounts of volatile metal compounds, and that neither of these documents disclosed or suggested the use of a catalyst comprising a composite oxide within the meaning of the patent in suit.

- VI. The appellants requested that the decision under appeal be set aside and European patent No. 0 323 195 be revoked.

The respondent requested that the appeal be dismissed

and the patent be maintained.

Reasons for the Decision

1. The appeals are admissible.
2. *Novelty*
 - 2.1.1 D1 discloses the selective removal of nitrogen oxides from waste gases using catalysts consisting essentially of vanadium oxide and molybdenum oxide and/or tungsten oxide. It is indicated that it is not clear in what oxidative states the vanadium, molybdenum and tungsten are contained in the catalyst and that the catalyst may further contain complex oxides of vanadium and molybdenum such as $\text{Mo}_6\text{V}_9\text{O}_{40}$. The catalyst may be composed of the catalyst components alone or it may be preferable to have the catalyst supported on conventional carriers. The conventional carriers may be activated alumina, α -alumina, silica gel, aluminosilicate, diatomaceous earth, silicon carbide, titanium oxide or the like (column 3, lines 7 to 24). The Board accepts the appellants' submission that a skilled person would consider the said complex oxide to be a composite oxide within the meaning of the patent in suit. In the examples of D1 neither such complex oxides nor a titanium oxide carrier have been used.
 - 2.1.2 Claim 1 of the patent in suit requires the use of a catalyst comprising in combination a composite oxide and titanium oxide. Such a specific combination is not disclosed in D1. The present combination can be considered to form a selection of a subgroup of the

generic group of oxides of vanadium and molybdenum with one selected member from the independent generic group of carriers. In agreement with the established jurisprudence on novelty, the Board holds that a specific combination of elements requiring the selection of elements from two known groups should be regarded as being novel; cf T 12/81, point 13 of the reasons, OJ EPO, 1982, 296.

2.1.3 The other jurisprudence cited by the appellants is not particularly relevant to the present case. The criteria for selection inventions mentioned in T 279/89 relate to the selection of a sub-range of a known broader range. This is not the case here. Two of the three criteria developed in T 279/89, ie the selected sub-range should be narrow and the selected sub-range should be far removed from the prior art preferences and examples, are meaningless in the present case where a choice is made by selecting elements from different groups of some length. The third criterion in T 279/89 that, the selected sub-range should not be arbitrarily chosen from the prior art but must be purposively selected, is in the Board's opinion not a proper novelty criterion but merely a confirmation of a previously formed opinion on novelty (see also T 666/89, OJ EPO 1993, 495, point 8 of the reasons). Moreover, as will be discussed in more detail in the inventive step evaluation, the present choice is not arbitrary.

2.1.4 T 164/92 (OJ EPO, 1995, 305) relates to the disclosure of a document comprising a computer program. The statement therein that the disclosure of a publication is determined by what knowledge and understanding can

and may be expected of the average skilled person in the technical field in question can be accepted. This, however, does not mean that any non-disclosed combination of disclosed entities that a skilled person can derive from a document is state of the art within the meaning of Article 54(1) EPC. Combinations, which are covered by generic definitions, can only be considered as forming part of the state of the art if the combination is made available to the public as a specific teaching with regard to a technical action (see also T 181/82, OJ EPO 1984, 401, point 8 of the reasons).

2.1.5 Likewise the appellants' argument that the present combination of entities would have been seriously contemplated by a skilled person and therefore lacks novelty, cannot be accepted. Apart from the question whether the criterion of "seriously contemplating" is a proper novelty criterion, there is no evidence that a skilled person would have seriously contemplated the use of the specified composition of matter in the claimed process.

2.2.1 The other novelty objection is based on Example XIV of D2. This example discloses a process for preparing a catalyst for the catalytic removal of nitrogen oxides from waste gases. An aqueous mixture of metatitanic acid, ammonium metavanadate and ammonium molybdate is dried, the dried mixture is pressed to tablets and the tablets are calcined at 500°C for 2 hours. The calcined catalyst comprises the elements Ti, V, Mo and O in a ratio falling within the ranges mentioned in present claim 1. The components in terms of chemical compounds present in the catalyst are not disclosed. The

reworking of this example confirmed that the said elements were present in the proportion as required by present claim 1 but the crystal structure was not revealed. The appellants admitted during oral proceedings that there was no proof that in the catalyst obtained according to said prior art example a composite oxide of Mo and V was present. They argued, however, that there was neither any proof that a composite oxide was present in the catalysts used in the process of the patent in suit and that the catalysts obtained according to the examples of the patent in suit fell within the realm of D2. The Board cannot accept this line of argument for a novelty attack.

If the claimed subject matter is not supported by the examples and cannot be obtained by a skilled person following the instructions in the description an objection under Article 83 EPC could arise. Opposition grounds under Article 83 EPC were not raised in the notices of opposition and are not at issue here.

In any case the Board sees no plausible reasons why the catalysts obtained according to the examples of the patent in suit would not contain composite oxides. The presence of these composite oxides after the first calcination step at 700°C is confirmed by X-ray diffraction and it is unlikely that by the following processing steps, taking place at lower temperatures (450°C), the composite oxides decompose or react with the titanium oxide carrier. The subject-matter of claim 1 is therefore supported by the description and novelty should be considered with respect to the subject matter as claimed.

2.2.2 The appellants' argument that, if the examples of the patent in suit result in a catalyst comprising composite oxides, the same must apply for the catalyst obtained according to said prior art example, cannot be accepted either. The process conditions for obtaining the composite oxides in the examples of the patent in suit are essentially different from those disclosed in Example XIV of D2. According to the patent in suit first a composite oxide is formed by calcining a grinded mixture of the oxides of Mo and V at 700°C for 2 hours. This composite oxide is then mixed with metatitanic acid, the obtained wet mixture is kneaded, the mixture is then dried and moulded and the moulded product is calcined in nitrogen at 450°C for 2 hours to obtain the catalyst (Example 1). It is unlikely that under the process conditions mentioned in D2, ie calcining the moulded product comprising metatitanic acid, ammonium metavanadate and ammonium molybdate at 500°C for 2 hours a composite oxide of Mo and V is formed in a measurable amount. The fact that according to D3 composite oxides of Mo and V are formed by calcining a dried mixture of ammonium paramolybdate and ammonium metavanadate at 560°C for 5 hours in air, does not prove the formation of a composite oxide in Example XIV of D2. Not only was the temperature lower and the calcination time shorter, the mixture in D2 contained titanium dioxide and was therefore not homogeneous. The presence of titanium dioxide might have hindered the formation of a composite oxide of Mo and V. The Board, therefore, concludes that there is not only no proof for the presence of composite oxides in Example XIV of D2 as confirmed by the appellants, but that there are even no plausible reasons for their presence. In the Board's judgement, the appellants'

submission that it is practically impossible to determine the presence of small amounts of composite oxides in a catalyst obtained according to the said example of D2, does not imply that the burden of proof shifts from the appellants to the respondent.

For these reasons the claimed process must be considered novel with respect to said prior art example.

3. *Inventive step*

3.1 According to the appellants D1 represents the closest prior art. The Board agrees that D1 is a suitable starting document for the inventive step analysis. D1 discloses that the process is suitable for the removal of nitrogen oxides from the waste gases exhausted from a boiler, heating furnace or internal combustion engine (column 1, lines 9 to 12 and column 4, lines 1 to 6). The examples of D1 show that catalysts comprising a mixture of vanadium oxide and molybdenum oxide on alumina or silica as carrier have a high conversion rate of nitrogen oxides (more than 95%) in gases comprising as pollutants NO and NO₂ eventually together with SO₂. In the patent in suit it is recognised that such catalysts are not deteriorated easily and exhibit a high denitration performance. It was however known that these catalysts were deteriorated when the waste gases contained volatile metal compounds such as the oxides of Se, Te, Tl and As as acknowledged in the patent in suit (page 3, lines 18 to 26). The problem underlying the invention was to reduce the deterioration of the catalyst when treating waste gases comprising substantial amounts of such volatile metal

compounds. The patent in suit proposes solving this problem by using in a process for removing nitrogen oxides from exhaust gases a catalyst comprising in combination titanium oxide and a composite oxide of vanadium and molybdenum. According to the appellants the performance of the catalyst used according to claim 1 was not any better than those of the catalysts used in D1 or D2. They pointed to the higher conversion rates mentioned in these document compared with the results of the examples of the patent in suit. The Board agrees that the conversion rates in D1 and D2 are higher but considers that the conditions under which they were measured were not comparable with the conditions used in the patent in suit. In the patent in suit measurements were performed with a gas containing high amounts of poisoning volatile metal compound (140 ppm As_2O_3) at a high space velocity ($120\,000\text{ h}^{-1}$) whereas in the examples of D1 and D2 the gas did not contain volatile metal compounds and the space velocity was at most 27400 h^{-1} (Example 7 of D1). In D1 it is explicitly disclosed that at space velocities above $100\,000\text{ h}^{-1}$ the conversion rate of nitrogen oxides is lowered (column 4, lines 43 to 51). From the comparative examples in the patent in suit, performed with the same volatile metal containing gas and the same high space velocity, it is apparent that in the durability test the catalyst comprising titanium dioxide and a composite oxide of V and Mo perform better (removal of at least 47% after durability test) than the catalyst comprising titanium dioxide and an oxide of Mo or V or an equivalent mixture of the oxides of Mo and V (removal of at most 37% after durability test). Although the comparative examples in the patent in suit are not exactly in conformity with any of the examples

of D1 or D2 there is no evidence or plausible reason why any of the prior art catalysts would have performed better than the catalysts of the comparative examples under the same test conditions. The Board is therefore satisfied that the process of claim 1 actually solves the above-stated problem.

3.2 D1 does not deal with the problem of deterioration of the catalysts in the presence of volatile metal compounds and there is no indication that catalysts comprising the composite oxide $\text{Mo}_6\text{V}_9\text{O}_{40}$ disclosed therein perform any better than mixtures of the oxides of Mo and V. Thus D1 does not contain any pointer to the claimed solution of the above-mentioned problem.

3.3 D2 is also silent about the said problem. Since it does not even teach the use of a composite oxide there is no hint towards the claimed solution.

3.4 D3 relates to studies on the heterogeneous oxidation of 1-butene over $\text{V}_2\text{O}_5\text{-MoO}_3$ catalysts. It discloses composite oxides of V and Mo but not in relation to the removal of nitrogen oxides from waste gases.

D4 relates to the removal of nitrogen oxides from waste gases and discloses the use of a catalyst comprising oxides of V and Mo but does not disclose composite oxides thereof and does not treat the problem of deterioration by volatile metal compounds.

3.5 Documents D5a, D6, D7 and D8 disclose that most fuels, in particular coal, contain small amounts of metals which can form volatile metal compounds such as As. D5 discloses that such volatile compounds are present in

the combustion gases of electricity works. D9 discloses that As is present in trace amounts all over the world. D10 discloses that coal can be used as fuel for boilers. The Board does not exclude that volatile metal compounds are present in the waste gases that can be treated by the processes disclosed in D1 or D2 but this has no impact on the novelty and inventive step of the use of the particular catalyst in the process as claimed in the patent in suit. There is thus no need to discuss these documents in more detail. Document EH2, mentioned in the contested decision, does not contain any pointer to the claimed solution. Since the parties did not rely on this document in the appeal proceedings there is no need to discuss this document here.

- 3.6 The Board therefore holds that the process of claim 1 is not only new but it also does not follow in an obvious manner from the state of the art and thus involves an inventive step within the meaning of Article 56 EPC. Claim 2 is dependent upon claim 1. Novelty and inventive step of the process of claim 2 follows from this dependency.

Order

For these reasons it is decided that:

The appeals are dismissed.

The Registrar:

The Chairman:

S. Hue

R. Spangenberg