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D E C I S I O N
of 10 June 2005

Case Number: T 0904/02 - 3.3.5

Application Number: 96916227.0

Publication Number: 0830201

IPC: B01D 53/94

Language of the proceedings: EN

Title of invention:

Diesel engine exhaust gas purification system

Patentee:

JOHNSON MATTHEY PUBLIC LIMITED COMPANY

Opponent:

Umicore AG & Co. KG

Headword:

Exhaust gas purification/JOHNSON MATTHEY

Relevant legal provisions:

EPC Art. 54, 56

Keyword:

"Novelty: yes, implicit disclosure not convincingly shown"
"Inventive step: yes"

Decisions cited:

-

Catchword:

-



Case Number: T 0904/02 - 3.3.5

D E C I S I O N
of the Technical Board of Appeal 3.3.5
of 10 June 2005

Appellant:
(Opponent)

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Representative:

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Respondent:

(Proprietor of the patent)

JOHNSON MATTHEY PUBLIC LIMITED COMPANY
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Representative:

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Decision under appeal:

Interlocutory decision of the Opposition
Division of the European Patent Office posted
19 June 2002 concerning maintenance of European
patent No. 0830201 in amended form.

Composition of the Board:

Chairman: M. M. Eberhard
Members: B. P. Czech
S. U. Hoffmann

Summary of Facts and Submissions

I. The appeal is from the decision of the opposition division posted 19 June 2002 concerning the maintenance of the European patent No. 0 830 201 in amended form.

II. The amended independent claim 1 on which the decision under appeal is based reads as follows:

"1. A process for the reduction of nitrogen oxides (NO_x) in an exhaust gas from light-duty diesel engines, which exhaust gas comprises unburnt hydrocarbon and NO_x components, during an engine operating cycle, which process comprises passing the exhaust gas over a lean NO_x catalyst incorporating an adsorbent comprising a non-metallised zeolite such that unburnt hydrocarbons in the exhaust gas are adsorbed onto the zeolite during those parts of the operating cycle having an exhaust gas temperature below 190°C and are desorbed from the zeolite during those parts of the operating cycle which result in exhaust gas temperatures from 190°C to 250°C, to give a combined and increased content of hydrocarbon components in the exhaust gas over the lean NO_x catalyst, and catalytically reducing at least a portion of the NO_x in the gases flowing from the engine to N₂ whilst simultaneously oxidising said combined, increased, amount of hydrocarbon."

III. As grounds of opposition, the opponents had invoked lack of novelty and lack of inventive step, relying *inter alia* on the following prior art documents:

D1A: US-A-5 354 720

D2: Engler, B. H. et al, "Catalytic Reduction of NOx with Hydrocarbons Under Lean Diesel Exhaust Gas Conditions", SAE Technical Paper Series: SAE 930735, International Congress and Exposition, Detroit, Michigan, March 1993

D3: WO-A-94/22564

The opposition division concluded that the claims as amended during the oral proceedings met the requirements of Article 123(2) and (3) EPC. It held that the process according to the amended claim 1 was novel over D3 and was also inventive over the cited prior art D1A to D3.

- IV. With its statement of the grounds of appeal, the appellant (opponent) maintained that the process according to the said amended claims still lacked novelty over D3. It also argued that the claimed process lacked inventive step in view of a combination of D2 and D3.
- V. In its reply, the respondent (proprietor of the patent) submitted that the appellant's arguments failed to make a case that the patent as amended lacked novelty and or an inventive step.
- VI. Oral proceedings took place on 10 June 2005.
- VII. The essential arguments of the parties can be summarised as follows:
According to the appellant, the claimed process lacked novelty over D3. D3 disclosed a catalyst as defined in amended claim 1, i.e. a catalyst for treating lean

exhaust gas from a light-duty diesel engine, and also comprising a non-metallised zeolite. Moreover, it was known from D3 that the content of NO_x was an issue in the treatment of diesel engine exhaust gas, even though this document was not focussed on this issue. D3 did not say what happened to the NO_x contained therein since at the filing time of D3 the NO_x content of the diesel exhaust gas was not critical. It was also known from D3 that the zeolite pores served to retain some of the gas-phase hydrocarbons ("HC") contained in the exhaust gas during start up or other periods when the catalyst is relatively cool and to release the hydrocarbons when the catalyst has been heated to higher temperatures. The temperature ranges defined in amended claim 1 were those which occurred under normal driving conditions. Zeolites would always statistically adsorb and desorb some molecules at the same time, hence there was no sharp temperature border involved between the adsorption and desorption stages. Since the same catalyst was used for treating the same exhaust gas under the same conditions, all that was contained in amended claim 1, and in particular the same NO_x conversion, would also inevitably occur according to D3. At the oral proceedings, the appellant emphasised that claim 1 only required that "a portion of the NO_x" was catalytically reduced, which portion could also consist of a few molecules only. Referring to the reactions shown in Table 2 of D2, and in particular to the reaction "CH₄ + 4 NO <=> CO₂ + 2 H₂O + 2 N₂" as an example for the reaction of a hydrocarbon with nitrogen oxide, it argued that some desorbed HC molecules would inevitably (or "by nature") statistically meet and catalytically react with the NO_x present in the exhaust gas when following the teaching of D3. Generally

speaking, D2 confirmed that where there was oxidation there was also reduction, and indicated that "the oxidation of the hydrocarbons has an important influence on NO_x reduction". The patent itself indicated that the known catalysts used with lean diesel engine exhaust gas both oxidised HC and carbon monoxide ("CO") and reduced NO_x. Similarly, NO_x reduction, improved due to the presence of the adsorbing zeolite, also occurred as an inevitable side effect according to D3.

Having regard to inventive step, the appellant argued at the oral proceedings that D3 represented the closest prior art since it also concerned the technical field of catalysts for diesel engines, was relevant for controlling NO_x emission and disclosed the use of platinum catalysts. D3 also disclosed a zeolite component on which HC were adsorbed at lower temperatures and then desorbed at the higher temperatures required for their reaction. Therefore, no structural change to the system was necessary. Starting from D3, the technical problem to be solved by the process of the patent in suit could be seen in "making use of the stored hydrocarbons in a further catalytic reaction". The process as claimed was obvious in view of D2 and the common general knowledge. D2 disclosed that it was necessary to increase the concentration of reducing agents in the exhaust stream to improve NO_x conversion and referred to the use of unburnt HC present in the exhaust gas as an elegant solution. D2 also taught that hydrocarbons were the most effective internal reducing agents to achieve NO_x conversion. Moreover, D2 illustrated the reactions, including the reduction of NO_x by HC, occurring inevitably (or "by nature") in such a system. The skilled person would

thus have realised that this reaction would also occur in the process according to D3, at least to a certain degree, involving at least a few molecules or a portion of the NO_x in the sense of present claim 1. Hence it was obvious to improve the catalytic reduction of NO_x by increasing the HC concentration as described in D2, using the HC desorbed from a non-metallised zeolite at 190°C to 250°C which is disclosed in D3.

The respondent argued that D3 concerned an entirely different application to the patent in suit. D3 did not relate to a lean NO_x catalyst and was not at all concerned with NO_x reduction. At the oral proceedings, it argued that D3 was primarily concerned with lowering the total particulate emission ("TPM") by catalytically oxidising the volatile organic fraction ("VOF") thereof, while a portion of the HC and CO contained in the exhaust gas may also be oxidised (page 8, lines 25 to 26). Referring to D1A, column 1, lines 52 to 62, it emphasised that the use of oxidation catalysts for diesel exhaust gases did not necessarily lead to a significant NO_x reduction. Hence, it was questionable whether the features of amended claim 1 could be considered as "inevitable results" in carrying out the process described in D3. It also referred to D2, sheet 7, to illustrate the different previous approaches to NO_x removal from lean diesel engine exhaust gases. The appellant considered D1A, and not D2 or D3, to represent the closest prior art. D3 disclosed catching HC on a zeolite component at temperatures which were too low for the catalytic oxidation thereof. Considering that D3 belonged to the different field of diesel oxidation catalysts and had nothing to do with NO_x reduction, the skilled person would not look at D3

when trying to improve NO_x emission. Only by applying ex-post facto considerations could D3 be considered to contain a pointer towards the claimed solution.

VIII. The appellant requested that the decision under appeal be set aside and that the patent be revoked.

The respondent requested that the appeal be dismissed.

Reasons for the Decision

1. The amended claims meet the requirements of Article 123(2) and (3) EPC. This was not disputed by the appellant.
2. The patent in suit concerns "improvements in the control of regulated emissions from diesel engines" (see section [0001]). More particularly, claim 1 is directed to a process for the reduction of NO_x in an exhaust gas from light-duty diesel engines. Considering this particular technical context, the board takes the view that a process encompassed by claim 1 must necessarily lead to a technically significant reduction of NO_x to N₂ and to a technically significant decrease of the amount of NO_x emitted with the exhaust gas over an engine operating cycle. This view is in conformity with what is indicated in the patent in suit (see paragraph [0003]).

Novelty

3. Document D3 generally relates to "a catalyst composition and a method for oxidizing oxidizable

components of diesel engine exhaust, in which at least some of a volatile organic fraction of the diesel exhaust is converted into innocuous materials, and in which gaseous HC and CO pollutants may also be similarly converted" (page 1, lines 11 to 15; page 3, lines 4 to 10). D3 primarily aims at providing a system for the further lowering of the TPM by oxidising at least some of the VOF contained therein. Although the oxidation catalysts of D3, "when employed as a diesel exhaust treatment catalyst, are effective for effectuating a reduction in total particulates, they are also capable, especially with the optional addition of platinum or other catalytic metal", "of providing the added advantage of also oxidising a portion of the HC and CO" (page 7, line 27 to page 8, line 27).

- 3.1 The oxidation catalyst according to D3 comprises a catalytic material comprising a mixture of high surface ceria and a zeolite. The catalytic material may optionally comprise high surface area alumina and may optionally carry a low loading of platinum or palladium catalytic metal dispersed thereon. Alternatively, or in addition, the zeolite of the catalytic compositions may be doped, e.g., ion-exchanged with a catalytic moiety such as one or more of hydrogen, platinum, copper, nickel, cobalt iron etc. As examples of specific applications for the method disclosed, D3 mentions treating the exhaust of a diesel-powered automobile or light truck. See in particular page 3, lines 10 to 24. D3 thus mentions the possibility of using a metal-free zeolite catalytic component. The board however notes that according to example 3 of D3, which is the only one which actually relates to the treatment of exhaust gases from light duty diesel engines (of 2.0 and 2.5

litre engines of passenger cars, see the paragraph bridging pages 27 and 28), the catalyst composition comprises a metal-doped Fe- β -zeolite.

- 3.2 As pointed out by the appellant, D3 refers to "U.S. Government limits" on diesel engine exhaust gas (page 8, lines 12, 13 and 16), as well as to "emissions measurements instrumentation and techniques" for HC, CO, TPM and **NO_x** (page 28, lines 4 to 7).

However, considering that the authors of D3 did not even report or comment on any NO_x concentration values measured, the quoted statements do not imply that a reduction of NO_x to N₂ must inevitably take place when performing a method according to D3.

- 3.3 According to D3, the zeolite serves to catalyse the oxidation of VOF and to crack larger VOF molecules. Moreover, the zeolite component of the catalyst is able to trap hydrocarbon molecules during periods when the exhaust gas is relatively cool, e.g. during start-up. The trapped hydrocarbons are believed to be either oxidised within the zeolite and/or released therefrom when the temperature of the catalyst is high enough to catalyse their oxidation. See in this connection page 2, last paragraph, the paragraph bridging pages 8 and 9, the paragraph bridging pages 10 and 11, and page 16, lines 23 to 27.

Considering that D3 is silent about the fate of the NO_x molecules present in the exhaust gas, the skilled person cannot directly and unambiguously derive from this document that the hydrocarbons adsorbed during cooler phases of the engine operation would actually

desorb at temperatures above 190°C and at the same time contribute significantly to a reduction of NO_x to N₂, rather than being oxidised by the available oxygen within or on the zeolite catalyst.

3.4 Moreover, D1A and D2 both contain information which makes it highly questionable whether the catalysts described in D3 as oxidation catalysts could generally be considered as "lean NO_x catalysts" in the sense of present claim 1.

3.4.1 D1A, a document relating to specific "lean NO_x catalysts" in the sense of the patent in suit, i.e. with catalysts achieving high conversion rates for HC, CO and NO_x, also contains a reference to prior art "diesel oxidation catalysts". The latter are stated to have high conversion rates for the oxidation of HC and CO. But it is also stated that they "do not alter the nitrogen oxide content in the exhaust gas", and that "a diminution in the nitrogen oxide content by reduction is difficult with these catalysts because of the high proportion of oxygen in the exhaust gas" (see D1A, column 1, lines 52 to 62).

3.4.2 In D2, the authors of this document, which include two of the inventors named in D1A, summarise the prior art in the field of NO_x removal from oxygen-rich exhaust gas of diesel engines. They conclude that there was (in 1993) still a necessity for the development of catalytic systems to remove HC, CO, particulates and NO_x simultaneously in oxygen-rich atmosphere overcoming the disadvantages of the mentioned prior art. Consequently, they aimed at providing a catalytic system which was able to fulfil future emission standards by decreasing

the exhaust gas CO-, HC-, NO_x- and particulate emission simultaneously (see in particular sheet 7, left-hand column, last paragraph to sheet 8, right-hand column, last paragraph).

3.5 As pointed out by the appellant at the oral proceedings, it was known that an overall reaction of hydrocarbons and nitrogen oxides to obtain carbon dioxide, water and molecular nitrogen was thermodynamically possible (see D2, sheet 7, section 1.2 and Table 2). However, D3 is silent about the reduction of NO_x to N₂ or any other reaction involving NO_x. Considering the number of different reactions and reaction mechanisms that may in principle occur (see sheets 3 and 5 of D2), it cannot be gathered from D3 which particular reactions between which molecular species would actually prevail apart from the overall oxidation of HC and CO when using the specific catalyst compositions according to D3, specifically in the exhaust gas from a light-duty diesel engine subjected to an operating cycle involving different loads and temperatures. Hence it is not directly and unambiguously derivable from D3 that a reduction of nitrogen oxides to molecular nitrogen inevitably occurs to a significant degree.

3.6 The board does not, at least for the sake of argument, exclude the possibility that in the process of D3 statistically and to a very small extent ("a few molecules") NO_x molecules present in the exhaust gas of D3 may actually react with and be reduced to N₂ by hydrocarbon molecules desorbed from the zeolite component of the catalyst during hotter phases of the engine operation, as alleged by the appellant. However, even if this indeed inherently occurred during

processes according to D3, as far as applied to light-duty diesel engines and relying on catalysts containing high-surface ceria and a non-metallised zeolite, D3 would still not be novelty-destroying. In particular, it has not been convincingly demonstrated by the appellant that the skilled person taking into account the common general knowledge would have understood that a process according to D3 would inevitably lead to a technically significant reduction of NO_x (i.e. of more than just a few molecules) to N₂ by desorbed hydrocarbons, although catalysts highly effective in the oxidation of HC and CO need not necessarily be effective in decreasing the concentration of the NO_x present (see points 3.4.1 and 3.4.2 above), and although none of the examples of D3 actually relates to the treatment of light-duty diesel engine exhaust gas with a catalyst comprising non-metallised zeolite. Neither has the appellant submitted experimental evidence conclusively showing that such a result would inevitably be obtained when treating light-duty diesel engine exhaust gas with a specific catalyst composition actually disclosed in D3 or a catalyst composition provided in accordance with the general teaching of D3.

- 3.7 Summarising, in the absence of convincing evidence, the board is not convinced that D3 represents a clear and unambiguous, explicit or implicit disclosure of a process according to present claim 1.
- 3.8 The appellant has not raised novelty objections on the basis of any of the other documents he cited in the appeal proceedings. The board also sees no reason for objecting to the novelty of the claimed process on the basis of any of these documents.

3.9 Inventive step

4. *Closest prior art*

4.1 Document D1A is concerned with the problem of reducing the quantity of NO_x in lean exhaust gas of diesel motor vehicle engines. As a solution to this problem, D1A proposes the use of specific catalysts. The catalysts disclosed have high conversion rates for CO, hydrocarbons and NO_x at exhaust gas temperatures as low as 225°C and comprise a first catalytic coating including alumina and/or ceria as a carrier for platinum and iridium, and a second catalytic coating of a specific zeolite. See in particular column 2, lines 53 to 59, column 2, line 65 to column 3, line 31 and claim 1.

4.2 Considering the similarity of the technical problems addressed and of the catalysts used according to D1A and the patent in suit, the board accepts that the process disclosed in D1A represents the closest prior art.

4.3 In contrast with what is required by claim 1 of the patent in suit, the zeolite to be used according to D1A contains copper and/or iron. The use of a non-metallised zeolite is not disclosed.

5. *Technical problem*

5.1 According to the patent in suit, the problem solved by the invention as claimed consists in the provision of a diesel engine system and process capable of a

substantial reduction in emissions of NO_x from light-duty diesel engines (see paragraph [0003]). At the oral proceedings, the respondent submitted that the technical problem to be solved starting from D1A consisted in improving passive lean NO_x conversion in diesel exhaust gas. It used the term passive to qualify those processes which did not rely on the additional injection of hydrocarbons.

5.2 It is plausible that by adding to a given lean NO_x catalyst component an adsorbent component capable of adsorbing HC during cooler phases of a cycle and releasing HC at temperatures above 190°C, more HC will be available for NO_x reduction to N₂ than in the absence of the adsorbent. Consequently, as confirmed by the test results shown e.g. in Figures 4a and 4b of the patent in suit, more NO_x will be reduced during those phases of the engine operating cycle when the desorbed HC combines with HC present in the exhaust gas. Consequently, the total NO_x emission over a cycle involving lower and higher temperatures will also be decreased as compared to the use of the lean NO_x catalyst in the absence of adsorbent. At the oral proceedings the appellant expressly accepted that a portion, but not all, of the NO_x was converted when carrying out the claimed process.

5.3 The respondent did not provide comparative data demonstrating an improvement, in terms of the absolute NO_x reduction actually obtained, over the results achievable according to D1A. In the absence of such data, the technical problem solved by the claimed invention can however be seen in the provision of a further process for the reduction of the NO_x contained

in the lean exhaust gas of light-duty diesel engines, which leads to a high NO_x conversion. In the present case, a high conversion means a conversion which is improved, over an engine operating cycle, with respect to that obtainable with the catalyst not containing a non-metallised zeolite adsorbent.

- 5.4 Since this technical problem is credibly solved by the claimed process, it remains to be seen whether this solution is suggested by the prior art cited by the appellant.
6. Document D1A is silent about an adsorption of HC contained in the exhaust gas onto components of the catalyst. Moreover, the zeolite coating of the catalyst necessarily contains copper and/or iron to provide "for optimised catalytic activity for the conversion of the noxious substances carbon monoxide, hydrocarbons and nitrogen oxides", see column 3, lines 26 to 30. Hence D1A, taken alone, cannot suggest the concept of storing unburnt hydrocarbons on an adsorbent comprising a non-metallised zeolite during cooler phases of an engine operating cycle, and of desorbing them during warmer phases of the cycle in order to react them with NO_x, thereby improving the overall reduction of NO_x.
7. Document D2 investigates various aspects of the catalytic reduction of NO_x with hydrocarbons under lean diesel exhaust gas conditions (see title). The target of the work reported was to find a catalytic system for simultaneously decreasing CO, HC, NO_x and particulate emission simultaneously in the oxygen-rich exhaust gas of diesel vehicles (see sheet 8, right hand column). The catalysts tested are zeolite-containing coated

monolithic cordierite honeycombs which have been impregnated with salts of noble metals, for example Pt, and activated in air (see sheet 6, left-hand column).

7.1 According to the authors of D2, the maximum possible conversion of NO_x by the hydrocarbons present in the exhaust gas from passenger car diesel engines was limited. For this reason, they consider it necessary to increase the concentration of reducing agents in such an exhaust gas stream (see sheet 3, right hand column, header "1.3 ENGINEERING ASPECTS" to sheet 4, right-hand column, line 5), with hydrocarbons being the "*most effective internal reducing agents to achieve NO_x conversion*" (sheet 27, fourth paragraph). More particularly, in D2 two measures are mentioned which both lead to such higher hydrocarbons concentrations: i) the use of fuel injection systems modified for higher HC emissions or ii) the injection of hydrocarbons, such as diesel fuel or gasoline, upstream of the catalyst (see sheet 4, right-hand column, the last two sentences). In those catalyst performance tests that were carried out with real vehicle engine exhaust gases, the authors of D2 relied on the constant addition of 800 vppm gaseous hydrocarbons upstream of the catalyst (see sheet 6, right-hand column, last paragraph; sheet 9, Table 7; and sheets 25 and 26, section "5. RESULTS AND DISCUSSION - VEHICLE TESTING").

7.2 In contrast therewith, in the claimed process it is the amount of hydrocarbons adsorbed on the non-metallised zeolite component during cooler parts of the engine operating cycle that is used as additional source of reducing agent for increasing the NO_x conversion. The adsorption and desorption of hydrocarbons is dependent

on the exhaust gas temperature and requires no particular constructional changes to the fuel injection or the provision of means for injecting hydrocarbons upstream of the catalyst.

7.3 This different technical concept underlying the process according to the patent in suit is not addressed by D2. Although the authors of D2 considered "*the utilisation of the unburnt hydrocarbon present in the engine exhaust gas*" as an "*elegant solution*" for reducing NO_x emissions, the only method they actually suggest in this connection is based on a totally different approach since it requires a fuel injection system modified for higher HC emissions, see sheet 4, right-hand column).

7.4 Hence, D2 cannot suggest modifying the catalysts for diesel engine exhaust gases known from D1A by incorporating an adsorbent comprising a non-metallised zeolite, or by replacing the copper- and/or iron-containing zeolite considered as mandatory in D1A by a non-metallised zeolite, to solve the problem stated above.

8. Document D3 is not concerned with the reduction of NO_x at all and does not report any results in terms of the NO_x concentrations obtained when performing the processes disclosed. Therefore, the board is convinced that the skilled person looking for solutions to the stated technical problem, would not take D3 into consideration.

Moreover, for the reasons given above (see points 3 to 3.7), the board considers that a technically significant reduction of NO_x to N_2 is neither implicitly nor explicitly disclosed in D3. Therefore, even assuming the skilled person would consider D3, it would not - without knowing the patent in suit - find therein any incentive whatsoever to modify the lean NO_x catalyst of D1A in order to solve the stated technical problem, let alone by using a non-metallised zeolite.

9. As pointed out above, the authors of D2, aware of the fact that the NO_x -conversion by catalytic reduction can be increased by increasing the HC-concentration of the diesel engine exhaust gas, relied on the (active) addition of hydrocarbon as a reducing agent upstream of the catalyst, see sheet 25, left-hand column. Hence, assuming for the sake of argument that D2 was to be considered as the closest prior art despite this additional step of actively introducing hydrocarbon into the exhaust gas, and considering the disclosure of document D3 as established above by the board, the skilled person trying to provide a further process for the reduction of NO_x in diesel exhaust gas which leads to a high NO_x conversion would not, without the application of ex post facto considerations, envisage combining D2 with D3 at all. D3 does not disclose a technically significant reduction of NO_x to N_2 (see point 8 above). Therefore, in any case, D3 contains no incentive to radically modify the concept suggested by D2 by replacing the injection of additional hydrocarbon by the use of a non-metallised zeolite adsorbent as a temperature dependent source of hydrocarbon.

10. The board cannot accept the approach adopted by the appellant at the oral proceedings, namely considering D3 as the closest prior art. D3 is only concerned with removing VOF, HC and CO, and therefore belongs to the technical field of diesel oxidation catalysts. In contrast therewith, claim 1 is directed to a process for the decreasing the amount of NO_x in diesel engine exhaust gas by reduction of NO_x to N₂, with simultaneous oxidation of HC. Moreover, for the reasons given above in connection with the disclosures of D3 and D2, the board is not convinced that, as alleged by the appellant, starting from D3 the skilled person would realise that NO_x reduction also occurred in a process according to D3 and would therefore be led to also make use of the adsorbed HC in a further catalytic reaction in view of the teaching of D2. The appellant's attempt to formulate the technical problem starting from D3 is thus based on unproved assumptions and ex-post facto analysis and cannot, therefore, be employed for the examination of inventive step.

11. The other documents cited by the appellant contain no additional information which, in combination with the preceding documents, would point towards the process of claim 1.

12. Therefore, the process according to present claim 1 was not obvious to the skilled person having regard to the prior art relied upon by the appellant. Hence it also involves an inventive step. The patentability of claims 2 to 5 is supported by that of claim 1.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar:

The Chairman:

A. Wallrodt

M. Eberhard