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**D E C I S I O N**  
**of 4 November 1999**

**Case Number:** T 0727/96 - 3.2.2

**Application Number:** 88118737.1

**Publication Number:** 0315985

**IPC:** C22B 1/08

**Language of the proceedings:** EN

**Title of invention:**

Improved fluidized bed process for chlorinating titanium-containing material

**Patentee:**

E.I. Du Pont De Nemours and Company

**Opponent:**

Tioxide Group Limited

**Headword:**

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**Relevant legal provisions:**

EPC Art. 54, 56  
EPC R. 78(3), 84

**Keyword:**

"Novelty (yes)"  
"Inventive step (yes) after amendment"

**Decisions cited:**

T 0192/88, T 0296/87

**Catchword:**

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Boards of Appeal

Chambres de recours

Case Number: T 0727/96 - 3.2.2

**D E C I S I O N**  
**of the Technical Board of Appeal 3.2.2**  
**of 4 November 1999**

**Appellant:**  
(Opponent)

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**Representative:**

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**Respondent:**  
(Proprietor of the patent)

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**Representative:**

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**Decision under appeal:**

**Decision of the Opposition Division of the  
European Patent Office posted 10 June 1996  
rejecting the opposition filed against European  
patent No. 0 315 985 pursuant to Article 102(2)  
EPC.**

**Composition of the Board:**

**Chairman:** W. D. Weiß

**Members:** R. Ries  
J. C. M. De Preter

## Summary of Facts and Submissions

- I. European patent No. 0 315 985 was granted on 16 March 1994 on the basis of European patent application No. 88 118 737.1.
- II. The granted patent was opposed by the present appellants on the grounds that its subject matter lacked novelty and lacked an inventive step with respect to the state of the art (Articles 100(a), 54 and 56 EPC).
- III. With its decision posted on 10 June 1996 the Opposition Division held that the patent could be maintained in the form as granted and rejected the opposition. The following documents were considered in the opposition proceedings:
- D1: Fluidized Bed Chlorination of Titaniferous Slags and Ores, by E. C. Perkins et al., US Bureau of Mines Report of Investigations 6317, 1963, pages 1 to 13
- D2: US-A-3 960 704
- D3: GB-A-1 459 967
- D4: GB-A-0 759 724
- D5: Perry's Chemical Engineers' Handbook, 6th Edition, 1984, pages 9 to 6, 9 to 7
- D6: GB-A-0 893 067

- D7: US-A-3 848 051
  
- D8: Carbon Attrition during the Fluidized Combustion and Gasification of Coal, Report No. DE-FG22-81PC40796, 1985, pages 110, 111
  
- D9: US-A-4 310 495
  
- D10: GB-A-0 776 295
  
- D11: CRC Handbook of Chemistry and Physics, page B 163
  
- D12: ASTM D409 (Extract), page 230
  
- D13: Petroleum Refining, J. H. Gary an G. E. Handwerk, Marcel Dekker Inc., 1984, pages 54 to 75
  
- D14: Photographs of the Calcined Petroleum Needle, Sponge and Shot cokes, one page, submitted on 14 May 1996 by du Pont de Nemours
  
- D15: Table of Data for Sponge, Fluid and Shot cokes, one page, submitted on 14 May 1996 by du Pont de Nemours
  
- D16 to
  
- D18: Declarations of Mr H. H. Glaeser, dated 30 June 1989; 18 December 1989; and 28 July 1993
  
- D19: Declaration of Mr A. M. Doyle of 17 September 1993
  
- D20: Affidavit by Mr C. D. Hewitt dated 1 May 1996

IV. An appeal against this decision was filed on 5 August 1996 and the fee for appeal paid at the same time. The statement of grounds was submitted on 7 October 1996 in which the following further documents were referred to:

D21: US-A-2 948 587

D22: US-A-4 096 097

D23: Wendell E. Dunn: "High Temperature Chlorination of Titanium Bearing Minerals: Part IV", Metallurgical Transactions, June 1979, vol. 10B, pages 271 to 277

The appellants (opponents) further developed arguments that the extension of the original term for response requested by the patentee with letter of 3 June 1997 was unallowable under Rule 78(3) and Rule 84 EPC.

VI. In the official communication dated 7 March 1999, the Board, for the sake of clarity of technical terms, further referred to document

D24: Carbon and Coal Gasification, Science and Technology, José L. Figueiredo ed., Martius Nijhoff Publishers, 1986, pages 512 to 517

VII. Oral proceedings were held before the Board on 4 November 1999. At the oral proceedings the patentees submitted, in substitution for all earlier requests,

- a revised set of claims 1 to 8 as a main request,

- a revised set of claims 1 to 8 as a first

auxiliary request,

and a description suitably adapted thereto.

Claim 1 of the main request reads as follows:

"1. A fluidized bed process for chlorinating titanium-containing material in which the titanium containing material is reacted with chlorine and a carbonaceous material, characterized in that the carbonaceous material consists of calcined petroleum shot coke."

The appellants (opponents) requested that the decision under appeal be set aside and the patent be revoked. An objection under Rules 78(3) and 84 raised in the written proceedings was no longer maintained at the oral proceedings.

The respondents (patentees) requested that the decision under appeal be set aside and that the patent be maintained in amended form according to the main request or the auxiliary request submitted at the oral proceedings.

VIII. The arguments put forward by the appellants can be summarized as follows:

In document D1, titaniferous materials are reacted with carbon in a fluidized bed chlorination process. As set out on page 3, last paragraph, a closely sized calcined petroleum coke was used as a reductant. Such a chlorination process is also disclosed in document D21. As set out in document D24, page 513, point 2.5,



Nomenclature, "petroleum coke" is a generic term for all special petroleum coke products such as green (raw) and calcined products. The delayed coker process results in a mixture of three grades: shot coke, sponge coke and needle coke. Thus, having regard to decision T 192/88 according to which "the fact that a disclosure belonging to the state of the art is of generic character, e.g. relates to a group of chemical entities without specifying its members, does not mean that it cannot take away the novelty of a patent claim comprising the same disclosure", the term "calcined petroleum coke" in document D1 covers all these forms of coke, and the instruction to use calcined petroleum coke means using any form of such coke which is available. Shot coke was available before the priority date of the patent at issue - as is evident from document D13, page 58, lines 8 to 10 - and it would inevitably be calcined before the chlorination process. Hence, the subject matter of claim 1 of the main request lacks novelty with respect to document D1, or alternatively, with respect to document D21.

Even if novelty could be acknowledged, the claimed process lacks an inventive step. In the light of document D1 as closest prior art, the technical problem underlying the patent at issue is to prevent the degradation of coke on exposure to the fluidized bed chlorination process and thus minimizing the entrainment of coke "fines" with the exhaust gases. The first and obvious solution to this problem for a skilled person is to look at the type of coke rather than re-designing the reactor by adding filters, cyclones, static precipitators etc, as it is for instance proposed in document D7. Those skilled in the

art know, however, that the reductant coke suitable for the fluidized bed chlorination process must provide an excellent match in hardness, porosity, regular shape and low price. Such a suitable type of carbon, whereby "carbon" is construed as meaning a synonymous expression for coke, is described in document D4, page 2, lines 55 to 108, according to which the carbon should be hard, spherical, resistant to attrition by the turbulent action of the fluidized bed solid materials and of such a particle size, that it will remain uniformly distributed throughout the fluidized mass and will not be entrained to any extent in the outflowing stream with the gaseous reaction products. Having regard to these requirements, petroleum shot coke is the first choice: its spherical particles exhibit a high hardness, and its reactivity is similar to that of sponge coke. Moreover, it is common sense that spherical "shot" particles are less abraded in a turbulent flow than those of sponge coke which are softer, irregular-shaped and of uneven or serrated form. In addition, petroleum shot coke is cheaper than sponge coke. Thus having an alternative source of supply of coke which is cheaper and less liable to attrition is a strong and obvious indication to favour petroleum shot coke. Therefore, the claimed process does not involve an inventive step.

XI. The respondents argued as follows:

With respect to petroleum coke, documents D1 and D21 give only a generic disclosure rather than a specific type of petroleum coke. Following the approach taken in decision T 296/87, the specific selection of calcined petroleum shot coke is, therefore, not anticipated by

the technical teaching of D1 or D21.

Regarding the problem to be solved when starting from document D1 as the closest prior art, except for the documents D4 and D7, none of the remaining prior art even addresses this problem and, therefore, this prior art cannot be helpful in solving it. Document D7, however, advocates a solution totally different to that claimed in the disputed patent, whereas document D4 is concerned with a method of chlorinating zirconium silicate and merely specifies that the reductant "carbon" should be of such particle size that it will not be entrained to any extent in the outflowing stream of the gaseous products, However, "carbon" must not necessarily be interpreted to mean coke. This estimation is supported by document D4, page 2, lines 84 to 88, which describes a "carbon material" containing over 99% fixed carbon, whereas the carbon content of petroleum coke is in the range of 93.6 to 97.8 %, as is shown in D14. Furthermore, apart from the fact that no reliable hardness data for petroleum shot, sponge and needle coke are available, this property is only one feature, but not the determining factor for selecting the appropriate type of coke. The reactivity of the coke is the more essential point to look at if the chlorination process is to be run effectively and economically. It is true that the delayed coke process generally results in a mixture of needle coke, sponge coke and shot coke. Since petroleum shot coke is generally said to be "unreactive and of less commercial value" compared to sponge or needle coke, (see document D24, page 515, point 3.2) it has never before been calcined like sponge and needle coke after separation but was discarded or used as a fuel for the cement

industry. Hence, there is no pointer in any of the cited documents leading a skilled person to the claimed solution, i.e. to decide on the selection of petroleum shot coke in substitution for conventionally used petroleum sponge coke.

## **Reasons for the Decision**

1. The appeal is admissible.

2. *Amendments*

In comparison with claim 1 as granted, each of the independent claims 1 of the main request and of the auxiliary request have been restricted by incorporating the term "consists of calcined petroleum shot coke", thereby excluding other forms of petroleum coke such as sponge, needle or fluid coke from the claimed process. This restriction, which has not been objected to by the appellants, is amply supported by the original disclosure, e.g. the examples 1 and 2, runs B and C. Hence, there are no objections to these amendments under Articles 123(2) or (3) EPC.

3. *Novelty*

The appellant, relying on decision T 192/88 argued that the generic term "calcined petroleum coke" is prejudicial to the novelty of claim 1 which is specifically restricted to "calcined petroleum shot coke". In this decision, point 3.4, a generic term for a chemical structure was held to define a group of

chemical entities comprising eight sterically different individual **equivalent** chemical compounds. The present case, however, is substantially different from the case considered in the above cited decision for the following reasons. As can be seen from document D13 relating to the properties and uses of petroleum coke, in particular page 57, Table 1 and below, only the properties of calcined petroleum sponge and needle coke are mentioned, and the main use of petroleum sponge coke is seen amongst others in the manufacture of titanium dioxide. Document D13 goes on to state saying on page 58, second paragraph, that occasionally a third type of coke, called "petroleum shot coke" because of the clusters of shot-sized pellets, is produced unintentionally. This document which represents the general knowledge of the skilled practitioner, therefore, supports the respondent's position that shot coke, after separation of the three grades of coke resulting from the delayed cooking process, had been considered as "waste material" which had not been considered worth being subjected to a qualifying treatment like calcination. Thus, a skilled person, upon reading the generic term "calcined petroleum coke" in document D1, would exclusively have implied the use of calcined petroleum sponge coke or, possibly, of needle coke. Hence, the generic term "petroleum coke" used in document D1, (or alternatively in documents D3, D9, D21 or D23) is understood by a skilled person to relate only to two equivalent and appropriate types of coke (i.e. sponge and needle coke) rather than to three types of coke. Therefore, the term "calcined petroleum coke" does not anticipate the term "calcined petroleum shot coke" used in claim 1.

Consequently, the subject matter of claim 1 is novel.

4. *Inventive step*

Among the cited documents, only D1, D3, D9 and D21 explicitly disclose the use of "calcined petroleum coke" as a reductant in a process for chlorinating titaniferous material in a fluidized bed. However, document D21 is essentially concerned with "calcined fluid petroleum coke and document D9 is directed to a low temperature chlorination process, i.e. at a temperature below 800°C, whereas the reaction temperature in the claimed process according to the patent at issue is in the range of 900 to 1300°C. Given that document D1, compared to document D3, tries to avoid excessive bed losses due to gas transport by operating the furnace at a low flow rate (below 17 lb/hr chlorine), the Board concurs with the parties that document D1 represents the closest prior art. However, neither document D1 nor D3 deal with the problem of coke degradation into "fines" by mechanical or chemical action.

Starting from document D1 as the closest prior art the technical problem underlying the patent at issue, therefore, is to provide a process which reduces, in the turbulent flow of the fluidized bed, the degradation of coke and thus the formation of coke fines which are entrained in the exhaust gases.

The solution to this problem consists in the use of calcined petroleum shot coke in replacement of conventionally used calcined petroleum sponge coke. It is apparent from the examples given in the patent at

issue that the problem has been successfully solved by the claimed process, since the coke content in the amounts of entrained solids in the exhaust gases are low.

Among the cited prior art, only documents D4 and D7 address the problem of the entrainment of carbon particles with the exhaust gases in the fluidized bed chlorination process. The appellants, in this connection, particularly referred to document D4, page 2, lines 55 to 108. Apart from being concerned with the chlorination of zirconium silicate rather than titanium oxide, the passage in document D4, however, reflects the necessity of providing a narrowly ranged particle size of carbon, preferably 8 to 75 mesh, in order to have the carbon particles remain uniformly distributed throughout the fluidized mass and not be entrained to any extent in the outflowing stream. Although the carbon granules should be hard, spherical or granular and resistant to attrition by the turbulent action of the fluidized solid  $ZrSiO_4$  materials, the term "carbon" used in document D4 does not necessarily mean "coke", as alleged by the appellant. This interpretation is supported by the fact that the representative chemical analysis of the carbon material in D4 specifies "over 99% carbon", whereas the carbon content of petroleum coke is in the range of 95 to 97.8% (cf. document D14). Even if the skilled person upon reading document D4 had interpreted the term "carbon" as some sort of coke or even petroleum coke, he would not have been immediately led to "petroleum shot coke", because this type of coke was not regarded as being appropriate for the fluidized bed chlorination process due to its low quality and its low reactivity

compared to petroleum sponge (regular) coke or needle coke (cf. document D24, page 515, point 3.2). The patentee pointed out in this context that in the delayed coking process petroleum shot coke is produced unintentionally and thus represents an undesirable by-product which is normally avoided and separated from the regular sponge coke before calcining the coke. This statement was not challenged by the appellant, and it is supported by the technical information given in documents D13, page 58, second paragraph and D24, page 515, third paragraph.

Document D7 is the only citation which - like the disputed patent - specifically deals with the formation of a certain amount of finely divided material or "fines" as a result of the reactions occurring and of abrasion. These fines which are entrained with the gases leaving the reactor represent a substantial loss of valuable material and give rise to difficulties and abrasion in subsequent operations (cf. D7, column 1, lines 34 to 43). However, contrary to the solution claimed in the patent at issue, document D7 advocates for conducting the solid fines entrained with the reaction gases into a separator or fines recovery section, continuously separating them from the gas stream and passing them directly back into the reactor by gravity flow. No incentive whatsoever is given in document D7 so that a skilled person would be prompted to select a particular type of coke, as has been done in the disputed patent to minimize the formation of "carbon fines".

In conclusion, the solution to the technical problem in the present case was not obviously derivable by a



skilled person from the state of the art. Consequently, the subject matter of claim 1 of the main request involves an inventive step. The dependent claims 2 to 8 relate to preferred embodiments of the process described in claim 1 and are, therefore, supported by the main claim.

## 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 with the order to maintain the patent in amended form on the basis of  
  
claims 1 to 8 and the  
  
description pages 2, 2A, 3 to 6,  
  
all submitted as main request at the oral proceedings of 4 November 1999.

The Registrar:

The Chairman:

S. Fabiani

W. D. Weiß