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17

Aktenzeichen / Case Number / N^o du recours : T 392/86

Anmeldenummer / Filing No / N^o de la demande : 83 305 126.1

Veröffentlichungs-Nr. / Publication No / N^o de la publication : 0 103 449

Bezeichnung der Erfindung: Single particle multi-metallic reforming catalyst
Title of invention: and process of using the same.
Titre de l'invention :

Klassifikation / Classification / Classement : C 10 G 35/09

ENTSCHEIDUNG / DECISION
vom / of / du 1 February 1988

Anmelder / Applicant / Demandeur : Mobil Oil Corporation

Patentinhaber / Proprietor of the patent /
Titulaire du brevet :

Einsprechender / Opponent / Opposant :

Stichwort / Headword / Référence : Catalyst/Mobil

EPÜ / EPC / CBE Article 56

Kennwort / Keyword / Mot clé : "Inventive step - Return to concept regarded as
superseded - Surprising effect"

Leitsatz / Headnote / Sommaire

Europäisches
Patentamt

Beschwerdekammern

European Patent
Office

Boards of Appeal

Office européen
des brevets

Chambres de recours



Case Number : T 392/86

D E C I S I O N
of the Technical Board of Appeal 3.3.1
of 1 February 1988

Appellant : Mobil Oil Corporation
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New York, N.Y. 10017 (US)

Representative : Jones, Alan John et al,
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Decision under appeal : Decision of Examining Division 029
of the European Patent Office
dated 4 June 1986 refusing European
patent application No. 83 305 126.1
pursuant to Article 97(1) EPC

Composition of the Board :

Chairman : K.J.A. Jahn
Members : R.W. Andrews
C. Payraudeau

Summary of Facts and Submissions

I. European patent application No. 83 305 126.1, filed on 5 September 1983 and published on 21 March 1984 (publication No. 0 103 449) claiming priority of 13 September 1982 from a prior application filed in the United States of America, was refused by a decision of the Examining Division 029 of the European Patent Office dated 4 June 1986. The decision was based on Claims 1 to 10 filed 17 January 1986. The independent Claims 1 and 8 read as follows:

"1. A method for preparing a single particle multi-metallic catalyst composite which comprises the steps of contacting a porous refractory oxide support with gaseous carbon dioxide in an amount and for a pressure and time sufficient to substantially saturate the pores of the support with carbon dioxide and thereafter effecting impregnation of the carbon dioxide contacted support with platinum, rhenium and iridium such that the metals are substantially uniformly distributed throughout the support.

8. A reforming process which comprises contacting a reforming charge stock under reforming conditions with the catalyst composite prepared by the process of any one of Claims 1 to 7."

II. The stated ground for the refusal was that the subject-matter of Claims 1 to 7 did not involve an inventive step in the light of the disclosure in, inter alia:

- (2) FR-A-2 017 701
- (3) EP-A-0 036 703 and
- (4) US-A-2 884 382.

The Examining Division also considered that the use of the catalyst so prepared in catalytic reforming was also obvious in view of the disclosure in document (3).

- III. An appeal was lodged against this decision by a duly confirmed telex on 30 July 1986 and the appropriate fee paid in due time. In the statement of grounds of appeal filed on 25 September 1986 and subsequent letters the Appellants contended that the problem underlying the disputed application was to provide a reforming catalyst which produces a high grade of gasoline of high octane number over an extended period of time under conditions of high severity. This problem was solved by selecting the tri-metallic combination of platinum, rhenium and iridium and reversing the trend of the recent prior art by reverting to the superseded prior art practice of a single particle common support.
- IV. With the reply to a communication from the Board the Appellant submitted an amended statement of claim and an amended page 4 of the description. The present Claim 1 differs from that refused by the Examining Division in that the expression "such that the metals ... throughout the support" has been replaced by the expression "and calcining the resulting composite". The words "about" in Claims 2 and 3 and "contains" in Claim 9 have been deleted and the expression "a highly paraffinic naphtha containing" inserted after the word "stock" in Claim 9.
- V. The Appellant requests that the decision under appeal be set aside and a patent be granted on the basis of Claims 1 to 10 and amended page 4 of the description filed with his letter of 17 December 1987.

Reasons for the Decision

1. ~~The appeal complies with Articles 106 to 108 and Rule 64~~ EPC and is, therefore, admissible.

2. There are no formal objections to the current version of the statement of claim since it is adequately supported by the original disclosure. The phrase deleted in Claim 1 was redundant in the light of the paragraph bridging pages 3 and 4 of the description. The addition of the calcination step to Claim 1 is necessary, since it is essential for the solution of the technical problem underlying the application. This feature finds support in the first complete paragraph on page 8. The amendment to Claims 2, 3 and 9 brings these claims into agreement with the corresponding description on pages 4 and 10. Claims 4 to 8 and 10 correspond to Claims 9 to 13 and 15 respectively as originally filed.

3. The application relates to a process for preparing a single particle multimetallic catalyst composite. Such a process is known from document (3). This document discloses that a multimetallic catalyst composite comprising a platinum group metal and one or more metals from Groups VIIIA, VIIA and IVB of the Periodic Chart of the Elements on a common support are prepared by saturating the support with gaseous carbon dioxide, thereafter impregnating with the desired metal and calcining the resulting catalyst composite (cf. Claim 1).

However, the catalyst composites prepared in accordance with the teaching of this document were considered to be unsatisfactory with respect to their ability to catalyse the dehydrocyclization of normal paraffins to aromatics and in the yields of benzene thus obtained. Therefore, in the

light of the closest prior art the technical problem underlying the application-in-suit may be seen in providing a process for the preparation of catalyst composites with enhanced dehydrocyclization activity. Moreover, the enhanced activity is to be coupled with higher yields of benzene with low methane makes and the ability to produce high yields of gasoline of high octane number under conditions of high severity over an extended period of time.

According to the patent application this technical problem is essentially solved by preparing a single particle multimetallic catalyst composite by saturating the pores of a porous refractory oxide support with gaseous carbon dioxide, thereafter impregnating the support with platinum, rhenium and iridium and calcining the resulting catalyst composite.

From a comparison of Example 9 with Examples 11 and 14 (cf. Table 2 of the application) and Example 19 with Example 20 (cf. page 19) the Board is satisfied that technical problem underlying the application is plausibly solved.

4. After examination of the cited documents the Board agrees with the conclusion of the Examining Division that this technical teaching is not disclosed in any of them and the claimed subject-matter is, therefore, novel. Although the subject-matter of the present application falls within the broad general terms of document (3) the Board takes the view that the technical teaching of this document does not reveal the content of the present subject-matter since only the preparation of unimetallic catalysts (cf. second paragraph on page 4 and Example 5), bimetallic single

particle catalyst composites (cf. the first complete paragraphs on pages 4, 8 and 9, Examples 1 and 3 and Claim 6) and trimetallic dual particle catalyst composites (cf. Example 7) is specifically disclosed.

5. It still remains to be examined whether the requirement of inventive step is met by the claimed subject-matter.
- 5.1 Document (2) discloses a process for the preparation of single particle trimetallic catalyst composites by impregnating a porous solid support with platinum, iridium and rhenium and calcining the resulting composite (cf. page 5, lines 4 to 37 and Example 1). However, in this document there is no disclosure of the pretreatment of the porous support before impregnation with the metals with gaseous carbon dioxide. Nevertheless, the pretreatment of the supports of reforming catalysts with gaseous carbon dioxide to ensure the uniform distribution of the metal throughout the uniform distribution of the metal throughout the support is known from documents (3) and (4) (cf. (3) Claim 1 and the third paragraph on page 3; (4) Claim 1 and column 6, lines 28 to 31). Thus, there is no doubt that the skilled person could have pretreated the support of the catalyst known from document (2) with gaseous carbon dioxide. However, the proper question to ask is not whether the skilled person could have done so, but whether he would have done so in the expectation of some improvement or advantage (cf. T 2/83, Simethicone Tablet/Rider, OJ 1984, page 265, particular point 7 on page 274).
- 5.2 In seeking an answer to this question it should be borne in mind that the recent developments in the art have been towards the use of dual particle catalysts in the catalytic reforming of hydrocarbons. Thus, US-A-4 288 348 (5), published on 8 September 1981, describes a catalyst

composite made up of a mixture of two components, one component comprising platinum and rhenium on a support and the second component comprising iridium on a separate support, both supports having been pretreated with gaseous carbon dioxide before impregnation (cf. Claim 1 and Examples 1 to 8). US-A-4 264 475 (6), published on 28 April 1981, discloses a catalyst composite consisting of platinum on a support and iridium on a separate support, neither of the supports having been pretreated with gaseous carbon dioxide before impregnation (cf. Claim 1 and Example 1). Therefore, the Applicant has turned his back on recent developments in the art as represented by documents (5) and (6) and returned to a single particle multimetallic catalyst first disclosed in 1970 (cf. document (2)) incorporating a pretreatment process which has been known since 1959 (cf. document (4)). These considerations may be regarded as one indication for the presence of an inventive step for the claimed subject-matter of the application (cf. T 229/85, Etching process/Schmid, OJ 1987, page 237, particular point 7 on page 241 and T 165/85, reported in European Patent Office Reports, Volume 2(3), pages 125-130, 1987).

- 5.3 From the results obtained in the catalyst dehydrogenation/-dehydrocyclization screening tests it is clear that the pretreatment of the supports of single particle bimetallic and dual particle trimetallic catalysts with gaseous carbon dioxide leads to a decrease in the yield of benzene (compare in Table 2 of the application the results of Examples 11 and 14 with those of Examples 12 and 15 respectively). Therefore, the skilled person would not have expected that the pretreatment of the support of the single particle trimetallic catalysts known from document (2) with

gaseous carbon dioxide would solve the problem underlying the application-in-suit of simultaneously enhancing the dehydrocyclization activity of the catalyst and increasing the yield of benzene (cf. Examples 9 and 10 of the application).

5.4 Therefore, in the Board's judgement the subject-matter of the present Claim 1 involves an inventive step. Dependent Claims 2 to 7, which relate to preferred embodiments of Claim 1, derive their patentability from this claim.

5.5 Claims 8 to 10, which relate to the use of the catalysts prepared in accordance with the claimed process, are also allowable in view of the patentability of Claim 1.

6. Therefore, the Board cannot agree with the grounds and conclusions of the decision under appeal. However, the patent sought cannot be granted at present since the description has not been brought into agreement with the amended statement of claim.

Order

For the above reasons, it is decided that:

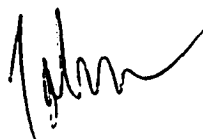
1. The decision under appeal is set aside.
2. The case is remitted to the Examining Division with the order to grant a patent on the basis of Claims 1 to 10 filed on 21 December 1987 and a description which has been brought into agreement with the amended statement of claim.

The Registrar:



F.Klein

The Chairman:



K.J.A. Jahn