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# DECISION
## of 31 July 1997

**Case Number:** T 0906/93 - 3.3.2

**Application Number:** 91201293.7

**Publication Number:** 0460738

**IPC:** B01J 27/16

**Language of the proceedings:** EN

**Title of invention:**
High surface area zirconia

**Applicant:**
Shell Internationale Research Maatschappij B.V.

**Opponent:**
-

**Headword:**
Zirconia/SHELL

**Relevant legal provisions:**
EPC Art. 54, 56

**Keyword:**
"Novelty (yes after amendment)"
"Inventive step (yes)"

**Decisions cited:**
-

**Catchword:**
-

Case Number: T 0906/93 - 3.3.2

# DECISION
of the Technical Board of Appeal 3.3.2
of 31 July 1997

**Appellant:**

Shell Internationale Research
Maatschappij B.V.
Carel van Bylandtlaan 30
2596 HR Den Haag (NL)

**Representative:** -

**Decision under appeal:** Decision of the Examining Division of the European Patent Office posted 28 May 1993 refusing European patent application No. 91 201 293.7 pursuant to Article 97(1) EPC.

**Composition of the Board:**

**Chairman:** P. A. M. Lançon
**Members:** M. M. Eberhard
J. H. Van Moer

## Summary of Facts and Submissions

I. European patent application No. 91 201 293.7 was refused by a decision of the Examining Division announced orally on 22 April 1993, with written reasoning posted on 28 May 1993. The decision was based on amended claims 1 to 13 filed on 21 January 1993.

II. The grounds for the refusal were lack of novelty for the product and use claims 10 and 13 and lack of inventive step for process claim 1.

The Examining Division held that the zirconia of claim 10 was not novel over the high surface area zirconias disclosed in US-A-4 440 875 (hereinafter D2), which had a surface area of 20 to 500 $m^2/g$. As it was well-known that calcination reduced the surface area, the value of 122 $m^2/g$ obtained in the example in D2 was inevitably lower than the values achieved in the application after calcination at 300°C for 1 hour. Therefore, it could not be assumed that the value of 122 $m^2/g$ was the highest that would be attained in D2 nor that the range 20-500 $m^2/g$ was speculative. No evidence had been submitted that the adsorption capacity of the claimed zirconia was higher than that of the zirconia disclosed in D2.

The Examining Division took the view that the process of claim 1 neither resulted in an unexpected effect nor offered an advantage over the process of D2. On the contrary, it was more complicated and led to a product which appeared to be identical to that of D2. Therefore, the aging step was merely arbitrary and did not involve an inventive step.

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III. The Appellants lodged an appeal against this decision. They filed an additional example (Appendix I) together with their statement of grounds of appeal and requested reimbursement of the appeal fee. In reply to a communication from the Board, the Appellants filed four sets of amended claims.

Oral proceedings took place on 31 July 1997. At the oral proceedings, the Appellants submitted four sets of amended claims as main request, first, second and third auxiliary requests respectively, in replacement of the previous requests. They also withdrew their request for reimbursement of the appeal fee. Claims 1, 10 and 11 of the main request filed at the oral proceedings read as follows:

"1. A process for the preparation of high surface area zirconia comprising aging zirconium hydroxide in the presence of an acid of an element of Group V or VI of the Periodic Table of Elements for at least 0.25 hours and calcining the zirconium material so-obtained."

"10. High surface area zirconia obtainable by a process according to any preceding claim, having a surface area of at least 230 $m^2/g$ after calcination at a temperature of 300°C."

"11. Use of a high surface area zirconia according to claim 10 as a catalyst or catalyst support in a process for the preparation of hydrocarbons from synthesis gas."

IV. Concerning the amended claims of the main request, the Appellants put forward *inter alia* the following arguments:

The range of specific surface area mentioned in D2 was purely speculative. A single example of a zirconium oxide catalyst having a surface area of 122 $m^2/g$ was not sufficient to provide support for the broad range of 20-500 $m^2/g$ mentioned in D2. A skilled person considering the relevant prior art, the information contained in the introduction of the present application and the disclosure of D2 itself, would have had considerable doubts as to the validity of the quoted range. The Appellant was entitled to the benefit of the doubt.

In any case, there was no enabling disclosure in D2 for the preparation of a zirconia having a surface area of 500 $m^2/g$. The additional example in Appendix I showed that the zirconia prepared following the instructions of D2 had a surface area of 254 $m^2/g$ after drying at 120°C. Other trials had been made, which all led to zirconias with a surface area of the same order of magnitude after drying. Therefore, a surface area of 500 $m^2/g$ after calcination at 300°C was clearly speculative. The additional example further proved that the zirconia prepared according to the process of D2 had a surface area of 210 $m^2/g$ after calcination at 300°C, ie lower than the claimed value of 230 $m^2/g$. Furthermore, the zirconias of the invention all exhibited a high level of surface area stability in contrast to the zirconia of D2 as shown in Figure 1 of Appendix I.

At the hearing, the question was discussed whether the zirconia disclosed in the example of D2, which had a surface area of 122 $m^2/g$ after calcination at 450°C, would exhibit a surface area of less than 230 $m^2/g$ after calcination at 300°C. The Appellants argued that the different curves in Figure 1 were not parallel to each other and had different profiles, some of them being concave and others convex with respect to the x-axis.

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Therefore, it could not be derived from Figure 1 that the zirconia of the example of D2 would have exhibited a surface area of at least 230 $m^2/g$ after calcination at 300°C. Furthermore, the value of 122 $m^2/g$ reported in D2 was that of a zirconia containing potassium oxide and not of zirconia alone. Citation FR-A-2 590 887 clearly taught that additives had an influence on the surface area of zirconia.

Concerning inventive step, it was in particular argued that the problem to be solved was to produce zirconia having a higher specific surface area and a higher surface area stability after calcination at high temperatures. D2 contained no information suggesting the use of an aging step in the preparation of zirconia in order to achieve zirconia having these improved properties.

V. The Appellant requested that the decision under appeal be set aside and that a patent be granted in the following version:

*Main request*: Amended claims and adapted description filed during the oral proceeedings, drawings as originally filed.

*Auxiliary requests*: Amended claims according to the first, second or third auxiliary request filed at the oral proceedings.

## Reasons for the Decision

1. The appeal is admissible.

2. The amended claims of the main request meet the requirement of Article 123(2) EPC. Claim 1 is based on the combination of claim 1 as filed with the statement on page 4 of the original description according to which a minimum duration of about 0.25 hours has to be observed for the aging step. The features of product claim 10 are disclosed in original claims 10 and 12. Claims 2 to 9 and 11 correspond to original claims 2 to 9 and 13 respectively.

3. The process according to claim 1 is new since none of the documents cited in the search report discloses aging the zirconium hydroxide in the presence of an acid of an element of Group V or VI in a process for the preparation of zirconia.

3.1 Concerning claim 10, it is observed that a product is not rendered novel by the mere fact that it is produced by a new process. Therefore, the question to consider is whether the cited documents disclose a zirconia having a surface area of at least 230 $m^2/g$ after calcination at 300°C.

D2 discloses a catalyst consisting of zirconium oxide promoted with at least one alkali metal compound. The zirconium oxide has a specific surface area in the range from 20 to 500 $m^2/g$ (see claims 1 and 2). A process for the preparation of this catalyst is given in the paragraph bridging columns 1 and 2. The zirconium hydroxide precipitated by addition of ammonia to a solution of zirconium oxychloride is separated by filtration, washed and calcined from 1 to 24 hours at a temperature in the range from 300 to 1000°C. The promoted catalyst is advantageously calcined before being used, suitably at a temperature in the range of 400 to 800°C (see column 2, lines 1 to 14). The sole example of D2 discloses a zirconium oxide catalyst containing 1% by weight potassium oxide and having a specific surface area of 122 $m^2/g$ after a previous calcination step at 450°C for two hours.

3.2 The upper limit disclosed in D2 for the surface area of the zirconia, ie 500 $m^2/g$, falls within the claimed range as it is implicit to the skilled person that the highest values of surface area can only be achieved with the lowest calcination temperature stated in D2, ie 300°C.

However, the Appellants argued that the upper limit of 500 $m^2/g$ was speculative and that D2 contained no "enabling" description for the preparation of such a zirconia.

D2 does not disclose parameters such as the concentration of the starting zirconium oxychloride solution, the ammonia concentration or the rate of addition of the ammonia. Therefore, the Appellants had themselves to choose these parameters. They pointed out at the oral proceeedings that they had performed several trials with different values of these parameters and obtained products which, after drying at 120°C, had a specific area of the same order of magnitude as in the additional example in Appendix I. The latter shows that calcination at 300°C for 1.5 hours reduces the surface area from 254 $m^2/g$ to 210 $m^2/g$. For the surface area of the calcined product to be of 500 $m^2/g$ after calcination at 300°C for at least one hour, the dried product must exhibit a surface area higher than 500 $m^2/g$. Even if, contrary to the Appellants' affirmation, it would have been possible by varying certain process parameters to produce products which, before calcination, have a surface area greater than 254 m²/g, for example about 300 m²/g, such values would still remain far removed from 500 m²/g, which is surprisingly high. In these circumstances, and in view of cited prior art itself and of the comments in the introduction of the present application, the Board has considerable doubts that the process described in D2 is disclosed in a manner sufficiently clear and complete for the skilled person to be able to obtain zirconias with a surface area of about 500 m²/g without undue burden. Therefore, in the absence of evidence to the contrary, the Board considered that the value of 500 m²/g indicated in D2 does not anticipate the claimed subject matter.

3.3 The additional example in Appendix I shows that a zirconia prepared by the process indicated in D2 exhibits a surface area of 210 m²/g after calcination at 300°C for 1.5 hours and that calcination at 400°C and 500°C leads to zirconias with surface areas of 141 m²/g and 59 m²/g respectively. Therefore, the resulting zirconia has a lower surface area after calcination at 300°C than the claimed zirconia. It is clearly derivable from these data that the general instructions given in D2 for the preparation of zirconia do not inevitably lead to a product having the claimed surface area when calcination is carried out at a temperature and for a duration both chosen in the lower part of the ranges indicated in D2.

3.4 At the oral proceedings the question was raised whether the zirconia prepared by the process of D2 and having a surface area of 122 m²/g after calcination at 450°C for 2 hours would exhibit a surface area of less than 230 m²/g after a calcination at 300°C. This question arose in view of the fact that, according to the interpolated curve E concerning the zirconia of the additional example (see Figure 1), this zirconia seemed to have a surface area slightly lower than 122 m$^2$/g after calcination at 450°C. The Board has decided to give the Appellants the benefit of the doubt in this respect for the following reasons. The value of 122 m$^2$/g disclosed in the example of D2 is not the surface area of zirconia alone but of the catalyst, ie zirconia promoted with 1 wt.% potassium oxide. It cannot be inferred from D2 whether or not the presence of the potassium compound during the calcination has an effect on the surface area of the final product. Furthermore, even if it were assumed that the potassium compound has no influence on the surface area, it still could not be directly and unambiguously derived from the curve E that a zirconia with a surface area of 122 m$^2$/g after calcination at 450°C for 2 hours would have a surface area of at least 230 m$^2$/g after calcination at 300°C. In view of the different profiles of the curves A, B, C, D and E in Figure 1, it cannot be predicted how the profile of a curve illustrating the stability of a zirconia as disclosed in the example in D2 would be, in particular whether such a curve would run parallel to curve E.

3.5 It follows from the above that the zirconia of claim 10 is considered to be new over the zirconia of D2. As none of the remaining documents cited in the search report discloses a zirconia having a surface area of at least 230 m$^2$/g after calcination at 300°C, the subject-matter of claim 10 meets the requirement of novelty. It results therefrom that the subject-matter of use claim 11 is also novel.

4. Turning to the issue of inventive step in connection with claim 10, the Board considers that D2 represents the closest prior art. Starting from this prior art, the problem underlying the application can be seen as

the provision of a zirconia having a higher surface area after calcination at 300°C and a higher surface area stability after calcination at higher temperatures.

According to amended claim 10 this problem is solved by a zirconia which exhibits a surface area of 230 $m^2/g$ after calcination at the said temperature and is obtainable by a process comprising aging zirconium hydroxide in the presence of an acid of an element of Group V or VI for at least 0.25 hours and calcining the resulting material. In view of the examples of the present application and of the additional comparative example of Appendix I, it is credible, in the absence of evidence to the contrary, that the said problem has been solved by the claimed product.

4.1 D2 mentions zirconias having a surface area of up to 500 $m^2/g$. However the process disclosed in D2 leads to zirconias with a surface area of less than 230 $m^2/g$ after calcination at 300°C (see preceding points concerning novelty). Zirconias having a higher surface area and a higher surface area stability at high temperature were desirable at the priority date, in particular for use as catalyst supports, but a process of preparation which makes it possible to achieve products with the desired higher surface area was not available. D2 does not contain any information suggesting that by aging zirconium hydroxide in the presence of an acid of an element of Group V or VI, it would have been possible to obtain zirconia exhibiting a higher surface area after calcination at 300°C and a higher surface area stability at higher temperatures.

4.2 FR-A-2 590 887 cited in the search report discloses that the obtention of a zirconia having the highest possible surface area is favourable to the dispersion of the catalytic phase and to its catalytic activity.

It deals in this context with the problem of stabilising the surface area of zirconia at high temperature (see page 1). This problem is solved by a composition containing zirconium oxide and an additive which is at least an oxide of elements selected in the group of silicium, the rare earths and aluminium (see page 1, last paragraph, and claim 1). The highest surface area obtained after calcination at 400°C is 90 $m^2/g$. After a further treatment at 550°C, the surface area is reduced to 85 $m^2/g$. There is no information in this document pointing to the claimed solution.

The remaining documents cited in the search report also do not contain any teaching which could give the skilled person an incentive to perform an aging step as defined in claim 10 in order to increase the specific surface area of zirconia to values of at least 230 $m^2/g$ after calcination at 300°C and to improve the surface area stability at higher temperatures.

4.3 For the preceding reasons, the product according to claim 10 fulfils the requirement of inventive step set out in Articles 52(1) and 56 EPC.

5. Since the product according to claim 10 meets the requirements of patentability, it follows that the use of the said product as defined in claim 11 is also patentable.

6. Inventive step of process claim 1 has to be considered independently of that of product claim 10, since claim 1 is directed to a process of manufacture of high surface area zirconia, ie zirconia which does not necessarily have a surface area of at least 230 $m^2/g$ after calcination at 300°C.

6.1 Starting from D2 as closest prior art, the problem to be solved by process claim 1 can be seen as the provision of a high surface area zirconia having an improved surface area stability after calcination at high temperatures (in particular 300 to 500°C). It is proposed that this problem be solved by the process defined in claim 1 which comprises an aging step in the presence of an acid of an element of Group V or VI for at least 0.25 hours. In view of the examples in the application and of the comparative example in Appendix I, the Board is satisfied that this problem has been solved by the claimed process.

6.2 From the considerations in points 4.1 to 4.3 above, it follows that the cited prior art does not suggest performing an aging step as defined in claim 1 in order to improve the surface area stability of high surface area zirconias after calcination at high temperatures.

Therefore, the process according to claim 1 also meets the requirements of patentability.

7. Claim 1 being allowable, the same applies to dependent claims 2 to 9 whose patentability is supported by that of claim 1.

## 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 grant a patent according to the main request: Amended claims of the main request and adapted description both filed during the appeal proceedings, drawings as originally filed.

The Registrar:

Martorana

P. Martorana

The Chairman:

P. A. M. Lançon

M.Es

JM.