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**Datasheet for the decision
of 12 August 2014**

Case Number: T 2284/12 - 3.3.06

Application Number: 03743238.2

Publication Number: 1478458

IPC: B01J21/04, B01J21/12,
B01J32/00, C07D301/10

Language of the proceedings: EN

Title of invention:

CATALYST CARRIERS

Applicant:

Saint-Gobain Ceramics and Plastics, Inc.

Headword:

Catalyst carriers/SAINT-GOBAIN

Relevant legal provisions:

EPC Art. 123(2), 82, 84, 54, 56, 52(1)

Keyword:

Amendments - allowable (yes)
Unity of invention - (yes)
Claims - clarity after amendment (yes)
Novelty - (yes) - non-disclosed combination of features
Inventive step - (yes) - unexpected improvement shown

Decisions cited:

Catchword:



**Beschwerdekammern
Boards of Appeal
Chambres de recours**

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Case Number: T 2284/12 - 3.3.06

D E C I S I O N
of Technical Board of Appeal 3.3.06
of 12 August 2014

Appellant: Saint-Gobain Ceramics and Plastics, Inc.
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted on 16 May 2012 refusing European patent application No. 03743238.2 pursuant to Article 97(2) EPC.**

Composition of the Board:

Chairman B. Czech
Members: E. Bendl
U. Lokys

Summary of Facts and Submissions

I. The appeal lies from the decision of the examining division to refuse the European patent application with the application number 03 743 238.2, published as EP 1 478 458.

II. In the contested decision the examining division found that the claims according to the then pending main request did not meet the requirement of unity of the invention (Article 82 EPC) and that the respective claims 1 of both then pending auxiliary requests did not meet the requirements of Article 123(2) EPC.

Moreover, the subject-matters of the respective claims 1 according to all then pending requests was considered to lack an inventive step. In this connection, the following documents were relied upon by the examining division:

D1 = EP 0 327 356 A1,
D2 = US 5 929 259 B1,
D3 = WO 97/46317 A1,
D4 = US 5 057 481 B1 and
D5 = US 5 380 697 B1.

III. Under cover of its statement of the grounds of appeal the appellant (applicant) filed four sets of amended claims as new requests, arguing that the amended claims overcame the objections that had led to the decision under appeal. The statement of grounds also includes an experimental report.

IV. In its communication dated 23 May 2014 the board gave a preliminary opinion, indicating that the new sets of claims did not appear to be objectionable under Article

82 EPC, but calling into question whether said amended claims complied with the requirements of Article 123(2) EPC, clarity and inventive step in the light of document D4.

- V. With its reply dated 11 July 2014 the appellant replaced three of the four sets of claims already on file by new sets and additionally held that the amended claims according to all the upheld requests were clear, met the requirement of Article 123(2) EPC, and that their subject-matter involved an inventive step.
- VI. Oral proceedings were held on 12 August 2014. The debate focused on the issues of clarity and inventive step. Thereupon, the appellant filed a fresh set of amended claims as sole request, in replacement of all its previous requests.
- VII. Claim 1 of this newly filed request reads as follows:

*"1. A carrier for a catalyst for the epoxidation of an olefin, the carrier obtainable by a method characterized by:
forming a mixture consisting of:*

- a) from 50 to 90 % by weight of a first particulate alpha alumina having an average particle size (d_{50}) of from 10 to 90 micrometers;*
- b) from 10 to 50 % by weight, based on the total alpha alumina weight, of a second particulate alpha alumina having an average particle size (d_{50}) of from 2 to 6 micrometers;*
- c) from 2 to 5 % by weight of an alumina hydrate;*
- d) from 0.2 to 0.8 % of an amorphous silica compound, measured as silica;*
- e) from 0.05 to 0.3 % of an alkali metal compound measured as the alkali metal oxide;*

and optionally conventional extrusion aids, burnout material, and/or water;
all percentages being based on the total alpha alumina content of the mixture, wherein d_{50} is measured after five minutes of sonification and represents the particle diameter at which there are equal volumes of particles larger and smaller than the stated average particle size;
and then forming the mixture into particles and firing the particles at a temperature of from 1250 to 1470°C to form the carrier;

wherein the carrier comprises at least 95 % alpha alumina, wherein the carrier has a surface area of from 1.0 to 2.6 m²/g, as determined by the BET method, and a water absorption of from 35 to 55 %, wherein water absorption is defined as the weight of the water that can be absorbed into the pores of the carrier as a percentage of the total weight of the carrier, the carrier further having pores which are distributed such that at least 70 % of the pore volume is in the form of pores having pore diameters from 0.2 to 10 micrometers and pores with diameters between 0.2 and 10 micrometers provide a volume of at least 0.27 mL/g of the carrier, wherein pore volume and pore size distribution are determined by mercury intrusion under pressures increased by degrees to a pressure of 3.0×10^8 Pa, using 130° contact angle and mercury with a surface tension of 0,473 N/m."

Dependent claims 2 to 16 are directed to preferred embodiments of said carrier.

VIII. The appellant requested that the decision under appeal be set aside and a patent be granted on the basis of

the claims according to the request submitted during oral proceedings.

IX. As relevant here, the arguments of the appellant put forward at the oral proceedings can be summarised as follows:

Claims wording

- Due to the amendments all issues under Article 84 and 123(2) EPC were resolved.

Inventive step

- D4 was the closest state of the art, in particular considering that carriers E and J of D4 were quite similar to the carrier of claim 1. However, considering the indicated pore volumes, their water retention value would be above the claimed limit of 55%. Moreover, they contained fluorides used in their preparation.
- By formulating claim 1 as a product-by-process claim requiring an initial mixture **consisting of** compounds (a) to (e), possibly together with some optional ingredients, the carriers as claimed were distinguishable from the ones described in D4.
- Fluorides, i.e. compounds present in some of the carriers disclosed in the prior art, were excluded by the wording of claim 1 ("*consisting*"), and were not encompassed by the terms "*extrusion aids*" or "*burnout materials*".
- According to the experimental report carriers resembling the ones of D4 as much as technically possible were prepared. This report demonstrated improved selectivities of the claimed catalyst/carrier combinations at lower temperatures, compared to the ones of D4 and D1.

- Considering that unexpected advantageous effects were demonstrated by means of the comparative tests, the claimed subject-matter involved an inventive step.

Reasons for the Decision

1. Admissibility of the amendments to the party's case

- 1.1 The amended claims according to the (sole) request at issue were filed during the oral proceedings.

The board accepts that they were filed in reaction to detailed comments and objections of the board that were debated in the course of the oral proceedings. The amendments made can be considered as an attempt of the appellant to overcome all pending objections. They did not raise new issues of particular complexity and contributed to the convergence of the debate.

- 1.2 The experimental report was filed with the statement of grounds of appeal to further corroborate the appellant's position that, contrary to the findings in the decision under appeal, the claimed subject-matter was indeed improved over the prior art and hence inventive.

- 1.3 Considering these particular circumstances, the board decided to admit the amended set of claims at issue as well as said experimental report into the proceedings despite their late filing (Articles 114(2) EPC and Articles 12(4) and 13(3) RPBA).

2. Allowability of the amendments - Article 123(2) EPC
 - 2.1 The board is satisfied that the amended claims are fairly based on the disclosure of the application as originally filed (published under the PCT as WO 03/072244 A1).
 - 2.1.1 In particular, amended claim 1 stems from a combination of product claim 1 and method claim 16 of the application as originally filed, wherein the raw material used for preparing the carrier is further strictly limited to a combination of compounds a) to e) and, optionally, extrusion aids, burnout material and/or water (basis: page 5, lines 4/5 of the published PCT application). More precise indications concerning the methods to be used for determining the parameter values "*average particle size d_{50}* " (basis: page 5, lines 10 to 13), "*water absorption*" (basis: page 4, lines 16/17), "*surface area*" (basis: page 3, lines 15/16) and "*pore volume and pore size distribution*" (basis: page 4, lines 7 to 11) have also been inserted.
 - 2.1.2 Claims 2 to 16 are dependent on claim 1 and represent preferred embodiments of the catalyst carrier claimed. They find a basis in claims 2 to 4, 6 to 13, 17, 20 to 22 of the application as originally filed.
 - 2.1.3 Therefore, in the Board's judgement, the amended claims at issue are not objectionable under Article 123(2) EPC.
3. Clarity of the claims - Article 84 EPC
 - 3.1 The appellant amended the wording of claim 1 by defining more precisely the raw material mixture used in the preparation of the carrier and by indicating in

more detail the measuring methods to be used for determining the values of the parameters characterising the catalyst carrier according to claim 1 (see point 2.1.1, *supra*).

3.2 Previously existing ambiguities as to whether (some of) the parameters mentioned in the claims characterise the starting compounds rather than the final carrier, were removed by way of the editorial amendments made.

3.3 The board is satisfied that the wording of the amended claims at issue is sufficiently clear and, hence, no longer objectionable under Article 84 EPC.

4. Admissibility of the "product-by-process" form of claim 1

4.1 The board accepts that limiting the claimed subject-matter to carriers with specific parameter values falling within the numerical ranges of the original product claim 1, which are made from compounds a) to e) according to the method of original claim 16, was not possible without also including the process features of original claim 16.

4.2 Moreover, by limiting the raw materials to be used in the preparation of the carrier to compounds a) to e) and optionally extrusion aids, burnout material and/or water, as required by claim 1 at issue, the use of fluoride is excluded. Thereby the claimed subject-matter is further delimited from the disclosure of the closest state of the art, i.e. D4 (see point 7.2, *infra*). Moreover, the subject-matter of product-by-process claim 1 at issue is clearly distinguished from the cited prior art by virtue of the amendments made (see also point 6, *infra*).

4.3 The board concludes that claim 1 is not objectionable for being (partially) put in a product-by-process form by virtue of the amendments made.

5. Unity of the invention

5.1 The set of claims at issue comprises claim 1 as the sole independent claim, with claims 2 to 16 being dependent therefrom.

5.2 Thus, the board is satisfied that the set of claims at issue is no longer objectionable under Article 82 EPC.

6. Novelty (Article 54(1), (2) EPC)

6.1 As already acknowledged in the communication dated 23 May 2014, the board is satisfied that the claimed carriers can be distinguished from the ones disclosed in the cited prior art.

6.2 In particular, the carriers of D1 are made using a fluoride (see claim 1 of D1), which possibility is excluded by the wording of claim 1 at issue. The carriers exemplified in D1 show higher water absorption values and different pore size distributions compared to the ones according to claim 1 at issue.

6.3 The carriers exemplified in D4 are not made from the same raw material mixture as the carriers defined in claim 1 at issue and also differ from the latter at least in terms of the surface area of the final carrier (carriers A to D have surface areas $< 1.0 \text{ m}^2/\text{g}$), in that they comprise a fluoride used in their preparation (carriers E to H, J to N) or in that no details are given concerning their pore size distribution (carrier I). Considering that carriers E and J of D4 have a pore

volume, measured by **water adsorption** or mercury porosimetry (D4: column 11, lines 21 to 25) of 0,68 and 0,668 ml/g, respectively, the board accepts that they have a water retention value greater than the upper limit of 55% specified in claim 1 at issue (see also the definition of water absorption on page 4, lines 16 to 19 as originally filed).

6.4 The board is also satisfied that none of the other prior art documents considered to be of relevance by the departments of first instance category "X" in the ISR and/or mentioned in the decision under appeal) discloses a carrier with all the features of claim 1 at issue. D2, D3 and D5 are silent on (*inter alia*) the use of alpha-alumina particles with an average particle size d_{50} in the range of from 10 to 90 micrometers and the pore size distribution of the carriers prepared.

6.5 Therefore, the board concludes that the subject-matter of claim 1 is new (Articles 52(1) and 54(1), (2) EPC).

7. Inventive step (Article 56 EPC)

7.1 The invention

7.1.1 The invention relates to ceramic catalyst carriers, more particularly to carriers for catalysts for the epoxidation of an olefin.

7.1.2 According to the application, epoxidation catalysts comprising a ceramic carrier according to the invention have excellent activity and unusually prolonged retention of a very high selectivity level at modest temperatures (page 2, lines 10 to 12 of the published PCT application).

7.2 Closest prior art

7.2.1 For the board, document D4 represents the closest prior art, since it *inter alia* focuses, like the patent in suit, on carriers for catalysts for the synthesis of ethylene oxide (column 1, lines 14/15), which exhibit high activity (column 9, lines 42 to 45) and high selectivities at both high and normal (i.e. lower) temperatures (column 9, lines 49 to 52).

7.2.2 More particularly, the exemplified carriers E and J of D4 (columns 20 and 21), contain at least 99 wt.% alpha-alumina and have a surface area, pore volume and pore size distribution as required by claim 1 at issue. Hence, they represent the most suitable starting point for the assessment of inventive step.

7.3 Technical problem

According to the appellant, the technical problem in the light of D4 (carriers E and J) consisted in the provision of improved ceramic catalyst carriers, which when used as carrier in a catalyst/carrier combination for the epoxidation of a polyolefin, show a very high selectivity level at modest temperatures (see also page 2, lines 10 to 12 of the published PCT application).

7.4 Solution

As a solution to the said technical problem, the application in suit proposes the carrier as defined in claim 1 at issue (see point VII, *supra*), which is characterised in particular

i) in that it is made by forming a mixture **consisting of** compounds (a) to (e) in specified relative amounts,

and optionally extrusion aids, burnout material and/or water, into particles and firing these at a temperature in the range of from 1250 to 1470°C, and

ii) by specific numerical ranges for the values of the parameters surface area, water absorption, pore diameter distribution and pore volume distribution of the carrier so obtained.

7.5 Success of the solution

7.5.1 In order to show that epoxidation catalysts comprising a carrier as claimed exhibit increased selectivities at lower temperatures than known catalyst/carrier combinations, the appellant referred to an experimental report included in its statement of grounds of appeal.

7.5.2 In this report, samples #5 and #6 represent carriers according to claim 1 at issue. The comparative carrier according to sample #3 of Table 4 is a reproduction sample supposed to be as similar as technically possible to carriers E and J of D4. Indeed, in the present case the board has no reason to doubt that the carrier of sample #3 is made according to the teaching of D4 and is thus representative of carriers as described in said examples E and J of D4. Sample #3 differs from the carriers according to claim 1 at issue essentially in that fluoride was used in its preparation and in that it has a water retention value of 68.8%, i.e. above the upper limit according to claim 1 at issue.

7.5.3 The board accepts that, as explained in the experimental report, the results shown in table 6 of the report give a clear indication that carriers as claimed achieve higher selectivities at lower

temperatures. Reference is made in particular to the following values:

#5 (invention): selectivity of 81.4 at 225 °C

#6 (invention): selectivity of 79.3 at 228 °C

compared to

#3 (as taught by D4): selectivity of 53.8 at 280 °C.

7.5.4 The board thus concludes that the appellant convincingly demonstrated that improved selectivities may be achieved at lower temperatures using a catalyst comprising a carrier according to claim 1 at issue instead of a carrier as disclosed by the closest prior art (examples E and J of D4) and that, hence, the technical problem posed is indeed effectively solved by the claimed carriers.

7.5.5 Although only one specific carrier prepared in accordance with the teaching of D4 was used for the comparison, the board has no reason to doubt that the improvement shown is achieved across the full ambit of claim 1.

7.6 Obviousness of the solution

7.6.1 It remains to be decided whether the claimed solution was obvious in the light of the state of the art.

7.6.2 Document D4

D4 generally discloses carriers defined in terms of physical parameters, said definitions overlapping to some extent with the definitions according to claim 1 at issue. Reference is made to the broad ranges of values suggested in D4 (see column 11, lines 10 to 29) as regards their BET surface area (less than 10 m²/g), pore volume as measured by mercury porosimetry (0.15 to

0.8 cm³/g) and median pore diameters (from 0.01 to 100 micrometers).

However, the specific carriers E and J of D4, which most closely resemble the carriers claimed, contain fluorides, which is something excluded by the wording of claim 1 at issue, and have water absorption (pore volume) values of 0.68 and 0.668 ml/g, respectively, i.e. outside the range according to claim 1 at issue.

It can neither be derived from D4 that the described properties of carriers E and J may also be achieved when omitting the use of fluorides in their preparation, let alone that the water absorption value of carriers so obtained could be as low as 0.55 in combination with a surface area value and a pore size distribution within the numerical ranges defined in claim 1 at issue.

Hence, nothing in D4 suggests to the skilled person seeking to solve the stated technical problem to modify the preparation method described in examples E and J such as to obtain carriers with all the combined features of claim 1 at issue.

7.6.3 The other prior art documents considered to be of relevance by the departments of first instance do not contain any more relevant information, let alone elements pointing towards the claimed solution of the technical problem.

7.7 Thus, the board concludes that the subject-matter of claim 1 and, consequently, of the subject-matters of dependent claims 2 to 16, are not obvious in the light of the state of the art, and therefore involve an inventive step (Articles 52(1) and 56 EPC).

Order

For these 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 the claims according to the request submitted during oral proceedings and a description to be adapted where appropriate.

The Registrar:

The Chairman:



G. Nachtigall

B. Czech

Decision electronically authenticated