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**Datasheet for the decision
of 12 May 2011**

Case Number: T 0673/07 - 3.3.07

Application Number: 01915393.1

Publication Number: 1268057

IPC: B01J 29/04

Language of the proceedings: EN

Title of invention:

Process for the production of a titanium silicalite shaped body

Patent Proprietors:

Evonik Degussa GmbH

Opponents:

BASF SE

Headword:

-

Relevant legal provisions:

EPC Art. 56
RPBA Art. 13

Relevant legal provisions (EPC 1973):

EPC Art. 54

Keyword:

"Late filed comparative test report - admissible (no)"
"Novelty (yes) - all requests"
"Inventive step (no) - arbitrary modification of a known process - Main Request, 1. and 2. Auxiliary Requests"
"Late filed claim request - admissible (no) - 3. Auxiliary Request"

Decisions cited:

-

Catchword:

-



Case Number: T 0673/07 - 3.3.07

DECISION
of the Technical Board of Appeal 3.3.07
of 12 May 2011

Appellants:
(Opponents)

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Global Intellectual Property
GVX-C006
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Representative:

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Respondents:
(Patent Proprietors)

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Decision under appeal:

Decision of the Opposition Division of the
European Patent Office posted 29 January 2007
rejecting the opposition filed against European
patent No. 1268057 pursuant to Article 102(2)
EPC.

Composition of the Board:

Chairman: J. Riolo
Members: G. Santavicca
P. Schmitz

Summary of Facts and Submissions

I. The appeal by the opponents lies from a decision rejecting their opposition against European patent 1 268 057.

II. The patent, granted on European patent application 01 915 393.1 (originating from international application PCT/EP01/03227 published as WO 01/72419 A1), comprised 14 claims, the independent claims reading as follows:

"1. Process for the production of a titanium silicalite shaped body by

a) forming a formable composition containing titanium silicalite, a binder and a pasting agent, so that the Curd curve of the formable composition has a plateau value in the range from 20 to 90 mm,

b) shaping the composition of step a) to form a green body,

c) optionally drying and

d) calcining the green body."

"12. Titanium silicalite shaped body obtainable by a process according to any one of the preceding claims."

"13. Process for the epoxidation of olefins with aqueous hydrogen peroxide in the presence of titanium silicalite shaped bodies according to claim 12."

"14. Process for the ammoximation of ketones with aqueous hydrogen peroxide and ammonia in the presence of titanium silicalite shaped bodies according to claim 12."

III. The patent was opposed on the grounds that the invention was insufficiently disclosed (Article 100(b) EPC) and lacked novelty and inventive step (Article 100(a) EPC) *inter alia* having regard to documents:

D1: WO-A-99/28029;

D2: WO-A-97/47614;

D3: WO-A-98/55229.

The patent proprietors *inter alia* submitted copy of a declaration (D19) by Dr Guido Stochniol, made before the USPTO in the proceedings concerning their parallel patent application, to show the plateau values and the Curd curves of some compositions illustrated by D3.

IV. According to the decision under appeal:

(a) the patent proprietors had proven that a Curd meter was publicly available at the Tokyo Metropolitan College, so the parameter feature of Claim 1 could be determined. Since the patent in suit contained examples and comparative examples and the opponents had not provided experimental evidence in support of their allegations, it had not been proven that the reproduction of the invention required an undue burden;

(b) As regards novelty and inventive step, the arguments of the opponents were either related to the assumption that it was not possible to obtain a Curd meter for determining the parameter of

Claim 1 or on plausibility considerations, without providing any item of evidence that the known processes for preparing shaped bodies of titanium silicalite (inherently) disclosed the plateau value of the Curd curve. Instead, the proprietors had provided comparative evidence (D19) over D3 and shown that the plateau value of the Curd curve was related to the lateral fracture resistance of the resulting shaped bodies. Since the examples showed that a plateau value of the Curd curve in the defined range resulted in titanium silicalite shaped bodies that were different from titanium silicalite shaped bodies the plateau value of which was outside that range, the claimed subject-matter was novel and inventive;

(c) Accordingly none of the grounds of insufficiency of the disclosure (Article 100(b) EPC) and lack of novelty and inventive step (Article 100(a) EPC) prejudiced the maintenance of the patent in suit.

V. In their statement setting out the grounds of appeal, the appellants enclosed a declaration of Dr Sadako Takasaki of 9 March 2007 (D22), visiting cards of Dr Takasaki and of Mr Takenaka (D23) and a declaration of Dr Erik Wassner of 9 January 2007 (D24).

VI. In response to the statement setting out the grounds of appeal, the patent proprietors (respondents) submitted a copy of an operation manual of the Curd meter Model M-301R, issued by the Firm Lio Electric Co., Ltd. (2-23-1, (1140) Yoyogi, Shibuya-Ku, Tokyo, Japan) (D25).

VII. In a communication of the Board in preparation for oral proceedings, in which the issues that needed debate and decision were indicated, the attention of the parties was *inter alia* drawn to two publications concerning the common general knowledge on Curd meters:

D26: Jun Toda et al., *Application of Principal Component Analysis to Food Texture Measurement*, presented at the 30th Annual Meeting of the Institute of Food Technologists, San Francisco, California, May, 1970, published in the Journal of Texture Studies 2(1971) 207-219; and,

D27: Lio, 1969, *Analysis of Curd Meter Measurement Curve*, Chori-Kagaku 2, 54 (in Japanese).

VIII. In response to the communication of the Board:

- (a) the appellants, with letter of 8 April 2011, submitted a copy of the Operator Manual of Curd meter-M-X ME-500 of the Firm Asuka Kiki, Tokyo, Japan, as Annex E1, and, as evidence supporting their arguments on lack of novelty, comparative test reports (Annexes E2 to E6) based, respectively, on Examples 5, 6 and 7 of the patent in suit and on Examples 2 of D3 and 5 of D2;
- (b) the respondents submitted two sets of amended claims as their first and second auxiliary requests (letter of 5 April 2011), then (letter dated 3 May 2011) commented on the latest items of evidence submitted by the appellants.

IX. Claims 1, 13 and 14 of the 1. Auxiliary Request filed with letter of 5 April 2011 were identical to Claims 1, 13 and 14 as granted (point II, *supra*). Claim 12 read as follows:

1. *Auxiliary request*

"12. Titanium silicalite shaped body obtainable by a process according to any one of ~~the preceding~~ Claims **5, 7, 8 and 9**".

X. The 2. Auxiliary Request submitted with letter of 5 April 2011 was made up of process claims 1 to 11 as granted (i.e. Claims 12 to 14 as granted had been cancelled). Thus, Claim 1 of the 2. Auxiliary Request was identical to Claim 1 as granted (point II, *supra*).

XI. Oral proceedings were held on 12 May 2011. The respondents handed over an "Eidstattliche Versicherung" (Affidavit) by Dr Ulrich Müller of 11 May 2011 (4 pages) as well as a set of amended Claims 1 to 8 as their 3. Auxiliary Request, Claim 1 reading as follows (compared to Claim 1 as granted, added features are indicated in bold):

"1. Process for the production of a titanium silicalite shaped body by

a) forming a formable composition containing titanium silicalite, a binder and a pasting agent, so that the Curd curve of the formable composition has a plateau value in the range from 20 to 90 mm,

b) shaping the composition of step a) to form a green body,

c) optionally drying and

d) calcining the green body, **wherein the binder is selected from aluminum oxide, silicon dioxide, hydrolizable silicon compounds and partial or complete hydrolysates thereof, boron compounds, phosphorous compounds, clay minerals and mixtures thereof and the pasting agent used is an aqueous medium optionally containing a water-miscible organic solvent."**

XII. The appellants essentially argued as follows:

Admissibility of the latest comparative examples

- (a) Since the decision under appeal found that the disclosure of the claimed invention was sufficient, the appellants bought a Curd meter and tried to use it according to the patent in suit. However, the use of the Curd meter on the basis of the patent in suit alone was not possible; staff needed to be trained by a Japanese expert and, during the crisis of the years 2009-2010, the project was delayed, then restarted as soon as possible. Given these circumstances, the experimental report filed with letter of 8 April 2011 (E2-E6) could not be submitted earlier. The "Eidestattliche Versicherung" (Affidavit) by Dr Ulrich Müller described how the experiments had been carried out, in order to reproduce the invention of the patent in suit. For reasons of economy and efficiency, some of the experiments were carried out with reduced amounts, however in such a way that the mechanical properties were not influenced thereby. As regards the kind of Curd meter used, it was not

contested that new and old Curd meters led to the same results. However, the new Curd meter no longer measured the plateau value, so it had to be calculated. As regards the shape of the curves, they did not differ from those illustrated e.g. by D19. In particular, the curve of E5 concerning Example 2 of D3 was highly relevant for novelty. In respect of the alleged prolongation of the proceedings, if the test report were admitted, the Board should also consider that the respondents were aware of the relevance of D3 since the beginning of the proceedings and, albeit possessing a Curd meter, had refrained from reproducing e.g. Example 2 of D3, despite the fact that this example was highly relevant for novelty and inventive step. Thus, the latest experiments should be admitted.

Lack of novelty

- (b) The steps defined in Claim 1 as granted were usual but the characterization of the formable composition by a plateau value in the range of 20 to 90 mm was not disclosed in the cited prior art. However, since D1, D2 and D3 illustrated formable compositions comprising titanium silicalite, pasting agents and binder, which were prepared in the preferred ranges of conditions as stated in the patent in suit, and which led to shaped bodies of comparable mechanical properties, the claimed process was not novel.

Closest prior art

- (c) D3, acknowledged in the patent in suit as disclosing a comparable process, described the closest prior art. D3 aimed at the preparation of titanium silicalite shaped bodies for use as catalysts for fixed beds having sufficient mechanical stability, so D3 aimed at the same objective as the patent in suit. The closest embodiment of D3 was illustrated by Example 2, its shaped body having very good mechanical properties. However, the process illustrated lacked any characterization of the plateau value.

Problem and solution

- (d) The patent in suit aimed at titanium silicalite shaped bodies having improved mechanical properties. No comparative tests over D3 were available. Example 4 and Comparative Example 4 of the patent in suit substantially differed from D3. In any case, even if Example 4 of the patent in suit were in compliance with Claim 1 of D3, it would not however correspond to the best example of D3, i.e. Example 2. As regards D19, which illustrated 2 Curd curves, concerning respectively Comparative Examples 1 and 3 of D3, it was not apparent why the examples of D3 had not been reproduced as well. Hence, the most relevant examples of D3 had not been reproduced to determine the plateau value of the formable composition. Since the problem stated in the patent in suit had not been solved, the problem solved over D3 was a further process for

the preparation of titanium silicalite shaped bodies.

Obviousness

- (e) The plateau value defined in Claim 1 as granted was a parameter feature of an intermediate product, the formable composition, not of the shaped bodies. Also, the determination of the plateau value was not a mandatory step of the process of Claim 1 as granted but could merely be the result of the choice of the starting materials. Hence, the definition of Claim 1 consisted of a desired presentation, which could not render inventive a known process.

Procedural matters - Non admissibility of a late filed claim request

- (f) The 3. Auxiliary Request was filed during the oral proceedings before the Board, thus too late. Having regard to D3 and the objections raised during the written proceedings, the request could have been submitted well before. So the 3. Auxiliary Request should not be admitted in the proceedings.

XIII. The respondents argued essentially as follows:

Non-admissibility of the latest comparative examples

- (a) The latest experimental evidence by the appellants had been submitted one month before the oral proceedings before the Board, i.e. too late for any possible verification by the respondents of what

had been done. No reasons whatsoever justifying that late filing had ever been provided by the appellants. In particular, the existence of a Curd meter was known since 2006 and the appellants had bought a Curd meter in 2008. The decision to stop training personnel was taken by the appellants themselves, so it was not clear why they needed so much time for carrying out the examples, let alone what difficulties were encountered and, if any, how they were overcome. Nevertheless, the experiments were not relevant, as no report describing how the reproduction of the examples had been carried out was submitted in time, so the reliability of the results could not be verified by the respondents before the oral proceedings. Also, the results were not plausible, as the plateau value had been calculated, not determined by the Curd meter. In that respect, the Curd meter M-301 of the respondents was completely mechanical, i.e. there was no software. Furthermore, the submitted curves, particularly those of E3, E4 and E6, differed markedly in their shape from those illustrated in the patent in suit, so it was apparent that the determination was not correct, or different curves (e.g. breaking curves) had been determined. Since, if the latest experiments were admitted, the respondents wanted to make further experiments to review those made by the appellants, which repetition required an adjournment of the oral proceedings and the continuation of the proceedings in writing, the late filed experiments E2 to E6 should not be admitted in the proceedings.

Novelty

- (b) There was no disclosure in the cited prior art of a formable composition having a plateau value in the range as defined in Claim 1 as granted. As regards the allegation that the plateau value was inherent in the processes of e.g. D1, D2 and D3, the onus of proof was on the appellants, who however had never proven beyond any reasonable doubts that the plateau value was inherently disclosed in any of said documents. In particular, neither the alleged disclosure of the preferred conditions of the claimed process nor the mechanical properties of the obtained shaped bodies were sufficient elements to prove lack of novelty. As regards the mechanical properties, it was known that calcination had a very strong influence on them, so simply prolonging calcination resulted in improved properties. Consequently, the plausibility arguments on lack of novelty were not convincing.

Closest prior art

- (c) D3 was the closest prior art document.

Problem and solution

- (d) The patent in suit contained comparative examples over D3, i.e. Comparative Example 4, which used the same binder of D3, albeit the teaching of D3 had not been exactly reproduced. Example 4 and Comparative Example 4 of the patent in suit showed that better mechanical properties were obtained by a formable composition fulfilling the requirement

of the plateau value over a formable composition not fulfilling the plateau value requirement, thus over D3. Hence, the problem stated in the patent in suit had been solved, so shaped bodies of improved mechanical properties were obtained from the claimed process.

Non obviousness

- (e) The plateau value as defined in Claim 1 as granted was not an unusual parameter but an unknown parameter. No such parameter had ever been disclosed for formable compositions of titanium silicalite. The plateau value was a quantification that served the purpose of keeping the formability of the composition within boundaries. It had never been acknowledged in the prior art that formability should be controlled in order to obtain better mechanical properties. Hence, the claimed process was not obvious. As regards the relation between mechanical properties and plateau values, D19 showed that no general relationship existed between them. Even if, for the sake of argument, the problem solved were the provision of a further process over D3, the process defined in Claim 1 as granted would not be obvious either, as the cited art does not mention the plateau value, which consequently could not be considered by the skilled person.

Procedural matters - Admissibility of a late filed claim request

(f) The 3. Auxiliary Request consisted of process claims only (i.e. the product claims were cancelled), whereby the additional features of Claims 4 and 10 had been incorporated into Claim 1. These amendments had a clear basis in the dependent claims, restricted the scope of granted Claim 1, i.e. did not extend the protection conferred by the granted patent. Therefore, the claim request was admissible.

XIV. The appellants (opponents) requested that the decision under appeal be set aside and the patent be revoked.

XV. The respondents (patent proprietors) requested that the appeal be dismissed, or, alternatively, that the decision under appeal be set aside and the patent be maintained on the basis of the 1. or 2. Auxiliary Requests, filed with letter of 5 April 2011, or the 3. Auxiliary Request filed during the oral proceedings before the Board.

Reasons for the Decision

1. The appeal is admissible.

Procedural matters - Amendment to a party's case - late filed evidence

2. The legal framework for the admissibility of *inter alia* late filed evidence is established by Article 13 RPBA,

which specifically deals with amendments to a party's case. In particular:

- (a) Article 13(1) RPBA generally specifies the principle of the Board's discretion for any amendments to a party's case after the filing of the grounds of appeal or any reply, whereby a non-exhaustive list of criteria for the exercise of the discretion is given, which includes the complexity of the new items submitted, the current state of the proceedings and the need for procedural economy.
- (b) Article 13(3) RPBA specifically deals with amendments sought after oral proceedings have been arranged, hence also during oral proceedings, and prescribes that the amendments should not be admitted if they raise issues which the Board or the other parties cannot reasonably be expected to consider without adjournment of the oral proceedings.

2.1 The latest comparative test reports by the appellants were submitted with letter of 8 April 2011 in reply to the communication by the Board in preparation for the oral proceedings, hence well after the submission of the statement setting out the grounds of appeal. Furthermore, a declaration by Dr Ulrich Müller, who was responsible for carrying out the comparative tests, providing information on how the examples of the patent in suit, D2, D3 as well as the characterization of the formed composition by means of a Curd Meter max ME 500 had been carried out, has been handed over only during the oral proceedings held on 12 May 2011.

- 2.2 In their letter of 3 May 2011, the respondents drew attention to the impossibility of verifying the correctness of the tests carried out by the appellants before the oral proceedings, as the data collection differed from that illustrated in the patent in suit and it was not apparent how the tests had been carried out.
- 2.3 The admission of the late filed test report is either unduly disadvantageous for the respondents, if no possibility of verification were conceded, or requires an adjournment of the oral proceedings and unduly prolongs the proceedings, if a possibility of verification were conceded.
- 2.4 Therefore, the late filed test report by the appellants cannot be admitted in the proceedings (Article 13 RPBA).

Main Request (patent as granted)

Novelty

3. It is not disputed that none of the cited documents mentions a formable composition containing titanium silicalite, a binder and a pasting agent having a Curd curve with a plateau value in the range from 20 to 90 mm, as defined in Claim 1 as granted of the patent in suit (point II, *supra*).
- 3.1 The patent proprietors, in a declaration by Dr Guido Stochniol (D19 in opposition appeal proceedings) (point 7 and relevant copy of the Curd curve) filed at the USPTO in the course of the examination proceedings of US-B2-6,849,570, have *inter alia* reproduced

Comparative Example 3 of US-B1-6,491,861 (a family member of D3). Comparative Example 3 of D3 illustrates a process of preparation of a titanium silicalite shaped body by

- (a) forming a formable composition containing titanium silicalite (120 g of powder), a binder (30 g of silica sol Ludox® AS-40) and a pasting agent (85 ml of water),
- (b) shaping (by a "Strangpresse", i.e. an extruder, to strands ("Stränge") the composition of step (a) to form a green body,
- (c) drying (at 120°C for 16 h) and
- (d) calcining (at 500°C for 5 h) the green body.

According to D19, the formable composition illustrated by Comparative Example 3 has a plateau value of 18 mm.

- 3.2 Also, since Claim 1 as granted defines the plateau value as a result to be achieved ("so that the Curd curve of the formable composition has a plateau value in the range from 20 to 90 mm"), it does not necessarily include a step of characterization of the formable composition before shaping, for determining whether or not its Curd curve has a plateau value in the range as defined. Hence, Claim 1 as granted encompasses a process that does not require a characterization of the formable composition to determine its plateau value, as acknowledged in the patent in suit (Paragraph [0011]), so step a) of

Claim 1 as granted merely defines a consequence of the choice of the components of the formable composition.

3.3 However, it has not been demonstrated by evidence that any of the known formable compositions, such as the further formable compositions of the examples of D3 (e.g. Example 2), (inherently) possesses a plateau values of 20 to 90 mm as defined in Claim 1 as granted.

3.4 So lack of novelty of the process of Claim 1 as granted has not been proven.

Closest prior art

4. The patent in suit concerns a process for the production of a titanium silicalite shaped body. A process for the production of *inter alia* titanium silicalite shaped bodies is known from D3, which, for all the parties, indisputably discloses the closest prior art. The Board has no reason to take a different position.

The disclosure of D3

4.1 According to D3 (page 1, lines 18, to page 2, line 29), abrasion-resistant mouldings comprising catalytically active materials were employed in many chemical processes, in particular in fixed beds. For their production, a binder, an organic viscosity-enhancing compound and a liquid for converting the material into a paste were generally added to the catalytically active material, i.e. a porous oxidic material, then the mixture was compacted in a mixing or kneading apparatus or an extruder. The resulting plastic

material was then moulded, e.g. in an extrusion press or extruder, and the obtained mouldings were finally dried and calcined. A number of inorganic compounds could be used as binders. Water was commonly used in the preparation of the mouldings, as a liquid for converting the material into a paste (pasting agent). Hence, the process steps defined in Claim 1 as granted were already known at the priority date of D3, i.e. well before the patent in suit.

- 4.2 Still according to D3 (page 3, lines 1-27), however, the known mouldings based on a porous oxidic material, e.g. titanium silicalite, had several disadvantages, such as insufficient mechanical strength for use as a catalyst in a fixed beds. In particular, secondary reactions of certain binders were undesirable, so whole classes of binders which could impart sufficient strength to such mouldings could not be used, e.g. because of other adverse properties. For example, aluminium-containing binders could not be used in the preparation of titanium silicalite which was used as a catalyst for the epoxidation of propylene with hydrogen peroxide, since the acidity induced by the aluminium-containing binder resulted in a greater degree of ring cleavage and formation of by-products. Moreover, titanium-containing binders could lead to high decomposition rates of the hydrogen peroxide used if these titanium-containing binders resulted in detectable titanium dioxide contents in the moulding. It was also undesirable to use binders which contained more than 100 ppm of alkali or alkaline earth metals. The catalytic activity of titanium silicalite could be severely adversely affected if the catalytically active

Ti centres were inactivated by the alkali metal or alkaline earth metal ions.

- 4.3 Thus, the object of D3 (paragraph bridging pages 3 and 4) was to provide a moulding which contained at least one porous oxidic material and had sufficient mechanical stability to be used as a catalyst in a fixed bed. When the moulding was used for catalytic reactions, the activity or selectivity losses due to secondary reactions of the added binder should be avoided in comparison with the prior art catalysts. A process for its production is also provided by D3. Therefore, D3 *inter alia* already addresses the object stated in the patent in suit (paragraph [0007]), i.e. a process for the production of titanium silicalite shaped bodies having improved mechanical properties.
- 4.4 In D3 (page 4, lines 8-16), this object is achieved by a moulding containing at least one porous oxidic material and exhibiting virtually no activity or selectivity losses when used as a catalyst, which can be obtained by use of a mixture containing at least one alcohol and water as a pasting agent in its production (The use of alcohol and water as a pasting agent is also contemplated in the patent in suit, as a particular embodiment, which is encompassed by the definition of Claim 10 as granted).
- 4.5 In particular, D3 *inter alia* discloses (Claims 2 to 8) a process for the manufacture of a shaped body containing at least one porous oxidic material such as titanium silicalite, which comprises the following stages:

(I) addition of a mixture containing at least one alcohol and water to a mixture containing a titanium silicalite and a tetraalkoxysilane or a mixture of two or more thereof, and
(II) kneading, moulding, drying and calcination of the mixture according to stage (I) after addition wherein an organic hydrophilic polymer or a mixture of two or more thereof is additionally added to the mixture in stage (I).

The alcohol in the mixture containing at least one alcohol and water can correspond to the alcohol in the tetraalkoxysilane (Claim 6).

The mouldings disclosed by D3 are suitably used for the epoxidation of organic compounds having at least one C-C double bond, for the hydroxylation of aromatic organic compounds, or for the conversion of alkanes to alcohols, ketones, aldehydes and acids (Claim 10), or for the epoxidation of an olefin, preferably for the preparation of propylene oxide starting from propylene and hydrogen peroxide (Claim 11). Thus, the mouldings of D3 are used as in the patent in suit.

D3 illustrates the preparation and properties of its mouldings, *inter alia* the one of Example 2, as follows: 120 g of titanium silicalite powder, synthesized according to Example 1, were mixed with 48 g of tetramethoxysilane for 2 h in a kneader. 6 g of Walocel (methylcellulose) were then added. For conversion into a paste, 77 ml of a water/methanol mixture containing 25% by weight of methanol were then added. The material obtained was compacted for a further 2 h in the kneader and then moulded in an extrusion press to give 2 mm

mouldings. The mouldings obtained were dried at 120°C for 16 h and then calcined at 500°C for 5 h. The lateral compressive strength of the resulting mouldings was tested and found to be 4.11 kg. 10 g of the mouldings thus obtained were processed to give chips (particle size 1-2 mm) and used as catalyst A in the epoxidation of propene with hydrogen peroxide, while maintaining activity and selectivity over a reference catalyst.

According to Comparative Example 1, if water were used as the pasting agent in Example 2, the lateral compressive strength of the resulting mouldings would be lowered (3.59 kg), whereas activity and selectivity would also decrease in comparison to the reference.

Problem and solution

5. The patent in suit, which *inter alia* acknowledges D3 as prior art, has, like D3, the object of modifying the existing manufacturing processes for the production of titanium silicalite shaped bodies in order to obtain shaped bodies having improved mechanical properties and being suitable for the epoxidation of olefins and the ammoximation of ketones (paragraph [0007]).
 - 5.1 According to the patent in suit (paragraph [0008], Claim 1), the object is achieved by a process for the production of a titanium silicalite body that comprises, in addition to the known (e.g. from D3, *supra*) steps of forming, shaping, drying and calcining the body, the requirement that the Curd curve of the formable composition has a plateau value in the range of from 20 to 90 mm.

5.2 Still according to the patent in suit (paragraph [0009]), if the Curd curve of the formable composition had a value as defined in Claim 1, the resulting titanium silicalite shaped bodies would have improved mechanical properties such as higher fracture resistance (determined by means of a pellet fracture resistance tester (TBH 28 Erweka) as described in paragraph [0042] of the patent in suit).

5.3 The patent in suit contains the following examples:

- (a) Reference Example 1, which merely deals with the preparation of titanium silicalite to be used;
- (b) Examples 1, 2 and 3 and Comparison Examples 1 and 2, in all of which aluminium oxide is used as binder;
- (c) Example 4 and Comparison Examples 3 and 4, in which a silicon dioxide precursor (TPA silicate solution) (obtained from reacting tetraethyl orthosilicate and tetra-n-propylammonium hydroxide in deionised water) (paragraph [0020] of the patent in suit) is used as binder, methylhydroxycellulose is used as shaping auxiliary (paragraph [0023]), water is used as pasting agent (paragraph [0027]), glacial acetic acid as a further additive (paragraph [0024]);
- (d) Examples 5 and 6 and Comparison Example 5, in which a silica sol is used as binder;
- (e) Example 7, in which additionally to Examples 5 and 6, boric acid is used as a further binder component.

5.4 The plateau value (mm) and lateral fracture resistance (N) determined for each formable composition and shaped body of each example (Ex n) and comparison example (CE n) are summarised in the following table:

Example	Ex 1	Ex 2	Ex3	CE 1	CE 2	Ex 4	CE 3	CE 4	Ex 5	CE 5	Ex 6	Ex 7
Plateau value	54 mm	81 mm	59 mm	94 mm	19 mm	33 mm	18 mm	14 mm	37 mm	14 mm	67 mm	45 mm
Lateral fracture resistance	87 N	46 N	103 N	7 N	19 N	32 N	could not be measured	10 N	21 N	15 N	21 N	57 N

5.5 It is apparent from the above that:

- (a) Examples 1 to 3 and Comparison Examples 1 and 2 all use aluminium oxide as binder, and shaped bodies having a high lateral fracture resistance are obtained from formable compositions having a plateau value within the range of Claim 1 as granted (paragraph [0055] of the patent in suit).
- (b) A comparison of Examples 5 and 7, in which comparable amounts of binders and other constituents are used, shows that the addition of boric acid (Example 7) as a binder component markedly increases the lateral fracture resistance (paragraph [0071] of the patent in suit).
- (c) In summary, the use of aluminium oxide and of the combination of silica sol and a boron compound as binder leads to the highest values for the lateral fracture resistance of the shaped bodies made thereby (paragraph [0071] of the patent in suit).

- (d) Example 4, Comparison Example 3 and Comparison Example 4 all use tetraethyl orthosilicate for forming the silicon dioxide precursor making the binder. According to the proprietors, these examples reflect the process disclosed by D3. However, water is used as the pasting agent, with the addition of glacial acetic acid, not a mixture of water and ethanol as taught by D3. Further, the non-measurable lateral fracture resistance of the shaped body of Comparison Example 3 is not the result of a lower plateau value (18 mm) alone but also of the use of a dried (instead of a calcined) titanium silicalite (this negative influence is also apparent from Example 2, if compared with Examples 1 and 3). Comparison Example 4 uses the same components of Example 4 but in bigger amounts and they are not processed (mixing and kneading) in exactly the same way as in Example 4, so the plastic properties of the formable composition of Comparison Example 4 are represented by a lower plateau value, and the lateral fracture resistance of the shaped body is inferior to that of Example 4. The lateral fracture resistance of the shaped body of Example 4 is lower than that of the shaped bodies of Examples 1-3 and 7. Thus, Example 4 and Comparison Example 4 only show that, if the same binder is used, the plastic properties of a formable composition play a role for obtaining shaped bodies of high lateral fracture resistance.
- (e) Even if it were acknowledged that Comparison Example 4 fulfils the conditions of Claim 1 of D3, the shaped body of Comparison Example 4 would still

not correspond to the specific shaped body obtained by the process illustrated by Example 2 of D3, which has a lateral fracture resistance of 4.11 kg (i.e. of about 40 N, as 1 kg_f or kp times 9.806₆₅ corresponds to 1 N). Lateral fracture resistances higher than that of Example 2 of D3 are obtained, in the examples of the patent in suit, only when aluminium oxide or silica sols and boron are used, which however are not defined in Claim 1.

- (f) Also, a plateau value that is slightly higher than the upper limit (90 mm) (i.e. 94 mm, Comparison Example 1) gives the lowest lateral fracture resistance determined. The use of dried (Example 2), instead of calcined (Examples 1 and 3), titanium silicalite leads to a higher plateau value, which however does not result in a higher lateral fracture resistance. According to the patent in suit (paragraph [0010]), a number of factors, *inter alia* the measuring conditions, affect the plateau value of the Curd curve. Also (paragraph [0034]), the calcination has a decisive influence on the mechanical stability. Hence, plateau value and mechanical stability do not necessarily depend on the same factors, which may be varied independently. Thus, the lateral fracture resistance does not only depend on the plastic properties of the formable composition alone. So no general relationship or correlation between the plateau value and the lateral fracture resistance (e.g. the higher the first, the higher the second) can be established.
- (g) Furthermore, a plateau value of 19 mm, as in Comparison Example 2, hence outside the claimed

range, gives a lateral fracture resistance of 19 N, which is comparable to that (21 N) obtained from a formable composition having a plateau value of 37 mm (Example 5) (i.e. within the claimed range), whereas a plateau value of 94 mm (slightly above the maximum defined in Claim 1 as granted) (Comparison Example 2) gives a very low lateral fracture resistance (7 N). So no lower and upper limits of the lateral fracture resistance corresponding to the lower and upper limits for the plateau values are generally definable.

- (h) Finally, the lack of any lower limit for the allegedly improved lateral fracture resistance is such that the lateral fracture resistance of the shaped bodies obtained from the process of Claim 1 as granted may well be lower than that of the known shaped bodies, e.g. of those illustrated by the examples of D3, such as Example 2.
- (i) Therefore, the range for the plateau values is arbitrary, at least as far as it is not related to the binder and the titanium silicalite used, nor to the calcination carried out.

5.6 Since the process of Claim 1 as granted is not restricted to the use of aluminium oxide as a binder, let alone to that of silica sol and boron, but it encompasses any binder, as well as any kind of titanium silicalite (dried or calcined) and pasting agents, and any calcination conditions as well, the alleged improvement in the mechanical properties of the shaped bodies obtained by the claimed process, as mentioned in

the patent in suit, is not necessarily attained over the whole breadth of Claim 1.

5.7 Even if Claim 1 were restricted to aluminium oxide, D3 mentions that its use as binder negatively affects the selectivity when the shaped body is used as catalyst in the epoxidation of propylene with hydrogen peroxide (page 3, lines 13-21), whilst the patent in suit is silent as to whether or not other properties of the catalyst (activity, selectivity) are maintained. Hence, it is not apparent whether e.g. activity or selectivity are affected by the choice of the binder, such as aluminium oxide, thus by the choice of the plateau value of the formable composition. Nor is it apparent whether, if any alleged improvement of the lateral fracture resistance were attained, activity and selectivity are maintained at a reasonable level, i.e. that the alleged improvement is not completely offset by disadvantages in other respects (Case Law of the Boards of Appeal, 6th edition, 2010, I.D.8.13).

5.8 It follows from the foregoing that there is no evidence on file showing, nor is it plausible, that the claimed process leads to shaped bodies having improved mechanical properties such as lateral fracture resistance over those disclosed by D3 over the whole breadth of Claim 1 as granted.

5.9 Therefore, the problem effectively solved is the provision of a further process suitable for the manufacture of titanium silicalite shaped bodies.

Obviousness

6. No prior art cited by the appellants mentions a process using a Curd meter in the manufacture of a titanium silicalite shaped body, let alone a formable composition for a titanium silicalite shaped body with a plateau value of a Curd curve in the range of Claim 1 as granted. Nor has it ever been shown by evidence that the formable compositions illustrated by D3 possess plastic properties quantified by a plateau value of the Curd curve as defined in Claim 1 as granted.
- 6.1 Nevertheless, as shown by the appellants (statement of grounds of appeal, points III.1.1.(b) and 1.1.3), the amounts and proportions of the components as well as the drying and calcination conditions illustrated by e.g. Example 2 of D3 fall within the (preferred) ranges disclosed by the patent in suit and lead to a product having high lateral fracture resistance. Hence, D3 suggests all of the known conditions of the process of Claim 1 as detailed in the patent in suit.
- 6.2 Also, as shown above (points 5), the plateau value of the Curd curve of Claim 1, i.e. the characterization, if any, of the plasticity of the formable composition (an intermediate product), does not necessarily contribute structurally to the lateral fracture resistance of the shaped bodies, which instead may well be influenced by other conditions such as calcination.
- 6.3 Consequently, the process of Claim 1 represents an arbitrary modification of the process of D3.

6.4 As established in the Case Law of the Boards of Appeal of the EPO (6th edition, 2010, I.D.8.19.6, in relation to T 939/92 (OJ 1996, 309)), when the technical problem is simply that of providing a further process of manufacture, as in the present case, an arbitrary choice from a number of possibilities, hence in the absence of a hint to do so, is not inventive if not justified by a hitherto unknown technical effect that distinguishes the claimed from other processes (e.g. D3). In the present case no unknown or surprising effect has been proven (points 5, *supra*).

6.5 It is not disputed that Curd meters were known before the priority date of the patent in suit, which fact is acknowledged in the patent in suit (paragraph [0012]).

6.6 Since the known Curd meters represented an obvious solution to the posed problem of merely proposing a further process of manufacture, their choice was devoid of any inventive character.

1. and 2. Auxiliary Requests

7. Since Claim 1 of the 1. and 2. Auxiliary Requests is identical, and identical to Claim 1 as granted (points VIII and IX, *supra*), the conclusions on the lack of an inventive step for the process of Claim 1 as granted apply *mutatis mutandis*. So the 1. and 2. Auxiliary Requests are not allowable.

3. Auxiliary Request

*Procedural matters - amendment to a party's case -
admissibility of a claim request filed at the oral proceedings
before the Board*

8. The 3. Auxiliary Request, filed during the oral proceedings before the Board, consists of only process claims, i.e. the product claims have been cancelled. In particular, Claim 1 results from the incorporation of the additional features of Claims 4 and 10 as granted in Claim 1 as granted.
- 8.1 The respondents justified the deletion of the product claims and the restriction of the process of Claim 1 as granted with the desire to provide a stronger limitation against the prior art, e.g. D3.
- 8.2 The Board, however, in a communication sent with the summons to oral proceedings, had indicated that the parties should be prepared to discuss D3 as the closest prior art, particularly its Example 2.
- 8.3 The opponent appellants cannot be expected to be prepared for any amendments to the respondents' claim requests which may be aimed at distinguishing them from D3. It is the patent proprietors' role to decide whether and when any auxiliary claim requests should be filed. The patent proprietors may have specific reasons to include or not to include certain subject-matter in their claims, which reasons are not always known to the opponents. So the opponents cannot usually be expected to prepare arguments against any unforeseeable claim requests that may be filed during oral proceedings.

8.4 Also, if the request were admissible, the additional features now included in Claim 1 do not appear to be suitable to impart any further distance from or distinction over D3, which in its Example 2 illustrates the use of tetramethoxysilane as a precursor of silicon dioxide as the binder as well as the use of a pasting agent comprising water and methanol. Hence, the 3. Auxiliary Request could only unduly prolong the discussion without any apparent possibility of changing the outcome as reached for the previous requests.

8.5 Therefore, the 3. Auxiliary Request is not admissible (Article 13 RPBA).

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.
2. The patent is revoked.

The Registrar:

The Chairman:

S. Sánchez Chiquero

J. Riolo