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Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 1113/97 - 3.2.2

Decision of the Technical Board of Appeal 3.2.2 of 25 April 2001 correcting errors in the minutes of the oral proceedings of 8 March pursuant to Rules 88 and 89 EPC

Appellant: (Proprietor of the patent)
Corning Glass Works
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Representative:
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Respondent: (Opponent)
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Decision under appeal: Decision of the Opposition Division of the European Patent Office posted 8 September 1997 revoking European patent No. 0 292 179 pursuant to Article 102(1) EPC.

Composition of the Board:

Chairman: W. D. Weiß
Members: R. Ries
J. C. M. De Preter

I. In the letter dated 27 March 2001, the appellant requested under Rule 88 EPC that the amended main request filed at the oral proceedings be corrected to include the drawings 1 and 2 of the patent as granted.

II. Given that the amended patent specification submitted at the oral proceedings includes references to the Figures and no reasons have been given to delete them, it is immediately evident that the drawings should remain part of the specification. The appellant's request is, therefore, allowable under Rule 88 EPC. Hence, the amended main request filed at the oral proceedings is corrected to include Figures 1 and 2 of the patent as granted.

III. In consequence thereof, the decision given by the Board in the minutes, point 2 on page 2 is completed by the words:

"and Figures 1 and 2 of the patent as granted"

The Registrar: The Chairman:

V. Commare W. D. Weiß

BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

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D E C I S I O N
of 8 March 2001

Case Number: T 1113/97 - 3.2.2

Application Number: 88304225.1

Publication Number: 0292179

IPC: C03B 8/02

Language of the proceedings: EN

Title of invention:
Method for producing ultra-high purity, optical quality, glass articles

Patentee:
Corning Glass Works

Opponent:
HERAEUS QUARZGLAS GmbH

Headword:
-

Relevant legal provisions:
EPC Art. 84, 123, 56

Keyword:
"Inventive step (yes) after amendment"

Decisions cited:
-

Catchword:
-

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Summary of Facts and Submissions

I. European patent No. 0 292 179 was granted on 12 October 1994 on the basis of European patent application No. 88 304 225.1.

II. The grant of the patent was opposed on the grounds that its subject matter did not involve an inventive step with respect to the state of the art (Article 100(a) EPC).

III. With its decision posted on 8 September 1997 the Opposition Division held that the claimed subject matter did not involve an inventive step and revoked the patent.

IV. On 7 November 1997 the patentee (appellant) lodged an appeal against the decision of the Opposition Division. The notice of appeal was followed by a statement of grounds submitted with letter (by telefax) of 19 January 1998.

V. With a letter dated 31 July 1998, the respondent submitted arguments that the claimed subject matter did not involve an inventive step in particular with respect to the technical teaching given by documents D1 and D4.

VI. Of the pre-published documents relied upon in the opposition proceedings, only the following have still been discussed on appeal:

D1: US-A-4 243 422

D3: Chemical Abstracts volume 105, No. 4, July 1986, page 288; Abstract No. 28665f & JP-A-61 58819

D3A: Translation into English of JP-A-61 58819

D4: EP-B-0 209 927

D5: Journal of Materials Science, volume 20, 1985, pages 4259 to 4297

D8: Journal of Non-Crystalline Solids, volume 42, 1980, pages 403 to 421

D11: EP-B-0 335 875

VIII. Oral proceedings were held before the Board on 8 March 2001 at the end of which the requests were as follows, taking into account the correction of the minutes according to the decision of the Technical Board of Appeal of 25 April 2001:

- The appellant (patentee) requested that the decision under appeal be set aside and the patent be maintained in amended form with the claims 1 to 19 and the description (pages 2 to 17) as submitted at the oral proceedings of 8 March 2001 and Figures 1 and 2 as granted.

- The respondent (opponent) requested that the appeal be dismissed.

Claim 1 as amended reads as follows:

"1. A method for producing fused silica granules comprising the steps of:

(a) preparing a solution which contains tetraethylorthosilicate;

(b) polymerizing the silicon in the solution with hydrochloric acid to form a SiO_2 gel;

(c) drying the gel at a rate which causes the gel to fragment into granules having a density of 1.29 - 1.59 g/cc, a pore size of $2x10^{-9}$ - $1x10^{-8}$ m, a surface area to mass ratio of 150 to 700 m^2/g and a mean particle size less than 1 millimeter; and

(d) sintering the granules at a temperature less than 1150°C to decrease the surface area to mass ratio of the granules to a value less than 0.5 m^2/g, the density of the granules after sintering being approximately equal to their maximum theoretically density."

IX. The arguments put forward by the appellant are summarized as follows:

The problem underlying the opposed patent is to provide fused silica granules for producing ultra-high purity optical quality glass. This problem is solved by the combination of technical features given in claim 1. The physical properties - defined in step (c) of claim 1 and depending essentially on the starting material tetraethylorthosilicate TEOS with hydrochloric acid as a catalyst and to some extent on the drying method - are pre-conditional for almost achieving the theoretical density of the granules by the "low temperature" sintering step below 1150°C which further minimizes the introduction of impurities through the furnace atmoshpere. The granulate obtained after step (d) ("artificial sand") exhibits a very low specific surface area of less than 0.5 m^2/g and is almost porous-free. Such a product cannot be obtained by using the conventional sol-gel process.

As opposed to the patent, document D1 neither discloses the necessity of using exclusively TEOS as the silicon containing organic compound nor the need to use hydrochloric acid as catalyst. On the contrary, the skilled reader is warned by document D1 against

introducing any chlorine into the process to prevent contamination and, consequently, formic acid is used as gelation agent in the examples. Moreover, document D1 fails to recognize the urgent need for keeping the physical properties of the fragmented gel within the claimed narrow limits to enable "low temperature" sintering as a follow-on which minimizes contamination of the glass particles by metals evaporating from the heating conductor coils of the furnace.

Example 4 given in document D3 and referred to by the opponent does not use TEOS as a starting material and, due to vigorous stirring the gel, a precipitation reaction in the sol is likely to occur. Document D5, on the other hand, is more concerned with the production a monolithic gel body rather than small particles of less than 1 mm as in the claimed method. Given this situation, the overall combination of technical features according to claim 1 which results in the high quality "ulta-pure" artificial sand is not derivable from any prior art referred to by the opponent. The claimed subject matter, therefore, involves an inventive step.

X. The respondent argued as follows:

Claim 1 comprises in step (c) a density of 1.29-1.59 g/cm^3. It is, however, unclear which type of density (e.g bulk density, true density, apparent density) is supposed to be meant, the more so, since the description of the patent fails to indicate any method for determining the density. Objection, therefore, arises under Article 84 EPC.

The closest prior art is reflected by document D1. With respect to steps (a) and (b) of the claimed method, this document discloses the use of orthosilicic acid esters of alkanols, especially C_1 and C_2 alkanols which

means in the case of C_2 ethylorthosilicate TEOS (cf. D1, column 3, lines 1 to 4). According to D1 column 3, line 66, the pH should be within the range of 2 to 4. Although formic acid is used in example 1 of D1 rather than hydrochloric acid as claimed, the wording in the description of the patent on page 5, line 31 and page 6, line 28 (.."an acid such as HCl..) makes clear that it is the amount of acid catalyst and not the type of acid which affects - in combination with other features - the physical properties of the gel particles. In this connection, it is apparent from example 4 given in document D3, that hydrochloric acid represents a typical catalyst for the sol-gel process.

As set out in document D1, column 4, lines 19 to 32, drying of the gel is best performed between 100 and 300°C and is continued until the silica gel achieves a free flowing powder. This means that the gel body is broken down into particles by the drying step. The physical parameters of the gel particles are said to be "typical" according to the patent at issue on page 6, line 29, when using TEOS, HCl and a specific TEOS:H_2O ratio. This is confirmed by document D5, page 4275, right hand column, paragraph 2, stating that after polymerization a porous structure of a gel exhibiting pores of 1 to 3 nm and total surface area of 500 to 1000 m^2/g is obtained. These data essentially comply with those mentioned in the patent.

Turning to step (d) of claim 1, document D1 discloses sintering of the particles at a temperature ranging from 1000 to 1400°C, preferably 1200°C (in the examples 1100°C) until "clear silicon dioxide granules" are formed (cf. D1, column 4, lines 55 to 68). Further reference is made in this context also to document D11, example 1, wherein after sintering a densification of the particles was achieved and the resulting powder product exhibited a BET surface area < 1 m^2/g indicating

that theoretical density has almost been achieved. Claim 1, therefore, does not comprise technical subject matter which involves an inventive step.

Reasons for the Decision

1. The appeal is admissible.

2. *Amendments*

In comparison with the granted claims, independent claim 1 has been restricted by:

(i) specifying in steps (a) and (b) the components of the solution (tetraethylorthosilicate, hydrochloric acid),

(ii) defining the physical properties of the gel granules in step (c) before sintering and

(iii) deleting the terms "approximately" and "about".

These restrictions, which have not been objected to by the respondent, are amply supported by original claim 2 in combination with page 5, lines 20/21 and 29/30 of the A-publication. Amendment (ii) has its basis on page 6, lines 27 to 30 and claim 10 as filed. The deletion of relative terms (iii) in claim 1, and in the dependent claims and the description of the patent at issue represents an admissible clarification to satisfy the requirements of Article 84 EPC. The description has been suitably adapted to the wording of the amended claims. Hence there is no formal objection to the present claims.

The respondents called into question the meaning of the term "density" as used in step (c) of claim 1. However, seen in the context of the claim and the specification as a whole, it is clear to the expert that - compared with the theoretical density after sintering - the values given in step (c) actually represent the "apparent density" of the granules rather than the true density (which is generally about 2 g/cm^3) or even the bulk density. Moreover, the expert skilled in this field of technology is aware of the appropriate methods which are to be used for determining the apparent density and, therefore, a specific reference in the patent to such a method is not necessary.

Hence, there are no objections to the amendments to the claims under Article 123(2), (3) and 84 EPC.

3. *The closest prior art*

The Board shares the view of the parties that document D1 reflects the closest state of the art. Like the patent at issue, document D1 relates to a sol-gel process for preparing granular quartz glass which can be melted to bubble free quartz glass for optical applications (cf. D1, column 5, lines 40 to 50). The hydrolysis of orthosilicic acid esters of C_1-C_2 alkanols, preferably tetramethylorthosilicate, is performed in water having a pH between 2 and 4. In order to prevent contamination by chloride ions, volatile organic acids, preferably formic acid or acetic acid are used (cf. column 1, lines 64 to 66; column 4, lines 1 and 2). The drying of the gel which is performed at slowly increasing temperatures in a range of 100°C and 300°C is achieved until the gel reaches a free flowing state (cf. D1, column 4 lines 18 to 23). Thereafter, sintering is carried out in a temperature range of 1000 to 1400°C, preferably 1200°C (cf. D1, column 4, line 51 to column 5, line 4).

4. *Novelty*

The subject matter of claim 1 is distinguished from the state of the art according to document D1 in that this document does not disclose the hydrolysis of TEOS together with hydrochloric acid as catalyst and in that it fails to mention the physical properties of the gel particles after drying.

None of the remaining documents referred to by the respondent discloses all the technical features given in claim 1. The subject matter of claim 1 is, therefore, novel. Given that novelty was not contested by the respondent, either in the opposition or in the appeal proceedings, there is no need to deal with this item in detail any further.

5. *Inventive step*

Starting from the technical teaching known from document D1, the problem underlying the patent at issue resides in providing an improved process for the production of ultra-pure fused silica granules ("artificial sand") having almost theoretical density and which can be further processed by powder pressing, slip casting etc. to produce green bodies.

The solution to this problem consists in producing granules fragmented from a gel body as an intermediate product, the granules having a density of approximately 1.29 to 1.59 g/cm^3, a pore size of $2x10^{-9}$ to $1x10^{-8}$ m, a surface area to mass ratio of 150 to 700 m^2/g and a mean particle size of less than 1 millimetre and sintering these granules at a temperature below 1150°C.

The central plank on which the opponent (respondent) has chosen to construct its case on inventive step is the set of premises that the claimed method lacks inventive step over a combination of the technical teaching given in documents D1 and D4.

However, document D1 does not disclose the essential features for solving the problem addressed in the patent at issue. To be specific, document D1 neither discloses the necessity of using TEOS nor the need to use hydrochloric acid as a gelling agent. On the contrary, given that chlorides are identified in document D1, column 1, lines 64 to 66, as being a source of contamination when producing high-purity granular quartz glass, volatile organic acids such as formic or acetic acid are used as a gelling catalyst rather than HCl. Since TEOS commonly is prepared from $SiCl_4$, it still may comprise chloride ions, and, therefore, tetra**methyl**orthosilicate instead of tetra**ethyl**orthosilicate (TEOS) is used in the examples 1 and 2 given in document D1. Thus, the skilled reader is advised to avoid any form of chloride contamination in the process when following the technical information given in document D1 and acting upon it. Hence, document D1 is clearly teaching away from the claimed method in which hydrochloric acid is chosen to be the preferred gelling catalyst.

Moreover, document D1 fails to mention the apparent density of the gel granules forming the free-flowing powder after drying, as mentioned in column 4, lines 18 to 22. It may be true - as alleged by the respondent - that the apparent density of the fragmented gel is "typical" for the process and that the artificial sand produced by the process according to D1 exhibits an apparent density which falls within the range given in claim 1 of the patent at issue. However, the

description of the disputed patent points out on page 6, lines 28 to 31, that the "typical" physical properties essentially depend on the H_2O-TEOS ratio (which according to the patent is preferably around 6:1), the amount of catalyst and the drying method. Since TEOS and HCl are excluded from the process according to document D1, it, therefore, remains questionable whether or not the "typical" properties featuring in claim 1 of the patent at issue are actually achieved. It is noted that the respondent did not contest this.

In view of these considerations, the evaluation of the technical features stipulated in document D1 is leading away from the claimed method and obviously does not allow a combination with other prior art which advocates the use of TEOS and specifically hydrochloric acid, as do documents D3, D4, D5 and D8.

In the process according to document D3, $CH_3OSi(OC_3H_7)_3$ is used instead of TEOS and hydrochloric acid constitutes the gelling agent. Even though the process disclosed in document D4 uses TEOS and HCl in a solution comprising water and ethanol, it seems to be more concerned with the production of a "suspension" rather than a gel (see in particular D4, examples 1 and 2). This is confirmed by the fact that after hydro-extraction, the precipitated solid material is re-dispersed and dried either by heating to 150°C (example 1) or by shock-freezing (example 2), both resulting in a powder. Consequently, the process according to D4 does not aim at producing a gel. Moreover, no evidence has been submitted by the respondent that the powder material obtained from the process according to documents D3 and D4 actually exhibits the properties defined in claim 1 of the patent at issue.

Turning to document D5, the hydrolysis of TEOS with HCl is disclosed in point 6.1.1.1, and pores having a diameter of 1 to 3 nanometres as well as a total surface area in the range of 500 to 1000 m^2/g are mentioned. The drying step according to document D5, however, aims at producing a **monolithic** high-porosity gel body rather than a gel broken up into granules of < 1 mm size. Even though the production of relatively small gel pieces, which could be sintered into glass, by controlled slow drying is also mentioned in document D5, this method nevertheless is rated as being impractical (cf. D5, point 6.1.1.2., lines 13 to 19). Consequently, document D5 dissuades from disintegrating the monolithic gel body into granules. The same technical information is given to the skilled reader in document D8, point 2.3, lines 4/5, stating that "the most serious problem [during the conversion of the gels to oxide glasses] is collapsing of the gels into pieces or powders on heating". Thus, according to the technical teaching given in documents D5 and D8, the drying step must be carefully controlled in order to produce an unfragmented monolithic gel body rather than small granules as claimed in the patent at issue.

D11 is even more remote in that it relates to a process using fume silica, a material which is definitively avoided in the method claimed in the patent.

In conclusion, there is no suggestion in any of documents D3 to D5, D8 and D11 relied upon by the respondent that the drying of the gel should be affirmatively used to produce granules having properties within specific ranges and to sinter these granules at temperatures below 1150°C to achieve almost theoretical density and to avoid contamination of the granules by substances contained the furnace atmosphere. Moreover, a combination of the approach to produce pure quartz glass given in document D1 with the

technical teaching disclosed in any of the remaining documents is - in the Board's view - not possible since D1 unambiguously dissuades the skilled reader form using any chloride containing compounds in the process.

6. Given this situation, the Board comes to the conclusion that the technical features forming the subject matter of claim 1 involves an inventive step. This is also true for the dependent claims 2 to 19 which relate to preferred embodiments of the process according to 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 maintain the patent in amended form with claims 1 to 19, the description (pages 2 to 17) as submitted at the oral proceedings of 8 March 2001, and Figures 1 and 2 of the patent as granted.

The Registrar: The Chairman:

V. Commare W. D. Weiß