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**Datasheet for the decision  
of 28 October 2010**

**Case Number:** T 0570/06 - 3.3.05

**Application Number:** 92913926.9

**Publication Number:** 0588950

**IPC:** C03C 17/30

**Language of the proceedings:** EN

**Title of invention:**

Method for treating fluoroaluminosilicate glass

**Patentee:**

MINNESOTA MINING AND MANUFACTURING COMPANY

**Opponents:**

THERA Patent GmbH & Co. KG  
Dentsply International Inc.  
Dental-Material Gesellschaft mbH

**Headword:**

Method for treating FAS glass/MINNESOTA MINING AND  
MANUFACTURING COMPANY

**Relevant legal provisions:**

EPC Art. 52(1), 56

**Keyword:**

"Inventive step (no; main request and first auxiliary request)  
- technical problem not solved over the whole scope of the  
claims"

"Inventive step (yes; second auxiliary request)  
- improvement credibly achieved by non-obvious method"

**Decisions cited:**

T 0223/05

**Catchword:**

-



Case Number: T 0570/06 - 3.3.05

**DECISION**  
of the Technical Board of Appeal 3.3.05  
of 28 October 2010

**Appellant 1:** MINNESOTA MINING AND MANUFACTURING COMPANY  
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**Decision under appeal:** Interlocutory decision of the Opposition  
Division of the European Patent Office posted  
16 February 2006 concerning maintenance of the  
European patent No. 0588950 in amended form.

**Composition of the Board:**

**Chairman:** B. Czech  
**Members:** E. Waeckerlin  
S. Hoffmann

## Summary of Facts and Submissions

I. The present appeals are from the interlocutory decision of the opposition division posted on 16 February 2006 concerning maintenance of European patent No. 0 588 950 in amended form.

II. Notices of opposition were filed by opponents 1, 2 and 3. All opponents requested the revocation of the patent on grounds of lack of novelty and lack of inventive step of the claimed method for treating fluoroaluminosilicate glass (hereinafter called "*FAS glass*"). The opponents relied *inter alia* on the following documents:

D1: DE 39 41 629 A1;

D1a: US 5 063 257 A (claiming priority from the same application JP 316303/88 as D1);

O1D2: US 3 066 112 A;

O1D3: DE 22 25 146 B2;

O1D4: Plueddemann, E.P.: Silane Coupling Agents; New York, London: Plenum Press, 1982, p. 32, 33, 49;

O1D5: Union Carbide® A-174 Silane Adhesion Promoter; New York: Union Carbide Corporation, January 1968;

O2D2: EP 0 318 153 B1;

O2D3: Stermann, S.; Mardsen, J.G.: Silane Coupling Agents; Industrial and Engineering Chemistry, vol. 58, no. 3, 1966, p. 33 - 37;

O3D4: EP 0 060 911 A1;

O3D5: WO 88/05652 A1.

III. By its first decision in the present case, posted on 27 January 2000, the opposition division revoked the patent. It acknowledged the novelty of the method as claimed in the patent as granted. However, starting from D1 as representing the closest prior art, the method according to all requests then on file was found to lack an inventive step, having regard to the disclosures of D1 and O1D5 or, alternatively, D1 and O1D3.

IV. The proprietor of the patent filed a first appeal against said decision of the opposition division. The reference number of the first appeal case was T 0184/00.

V. Opponent 1 withdrew its opposition with letter dated 12 February 2001.

VI. In its decision T 0184/00 of 21 November 2002, the board acknowledged the novelty of the method according to the amended independent claim 1 of the main request then on file over the disclosure of document D1. The wording of said claim 1 was as follows (features added to claim 1 of the patent as granted are highlighted by the board):

*"1. A method for treating fluoroaluminosilicate glass, comprising the steps of:  
a. mixing finely-divided fluoroaluminosilicate glass with an **acidic or basic** aqueous silanol solution, and  
b. drying the glass."*

The board remitted the case to the department of first instance for further prosecution.

- VII. Subsequently, the opposition division issued its second (interlocutory) decision dated 16 February 2006, which is the decision under appeal in the present proceedings. The subject-matter of claim 1, which was held novel in decision T 0184/00, was found to lack an inventive step in view of D1. On the other hand, the opposition division concluded that, having regard to the further limitation of using acidic or basic silanol solutions containing from 0.1 to 10 wt.% silanol and having a pH within specific ranges (as set out in claim 1 of auxiliary request 2 then on file), the claimed method satisfied the requirement of inventive step in view of the evidence on file.
- VIII. Appeals against said interlocutory decision were filed by the proprietor of the patent in suit (appellant 1), opponent 2 (appellant 2) and opponent 3 (appellant 3).
- IX. In its statement of grounds of appeal, appellant 1 maintained the main request refused by the opposition division, arguing that the claimed subject-matter was not obvious in view of the state of the art. Moreover, appellant 1 submitted a set of amended claims representing the first auxiliary request.

- X. In their respective statements of grounds of appeal, appellants 2 and 3 maintained that, taking D1 as the closest prior art and considering the state of the art, the experimental evidence on file and/or the common general knowledge, the claimed method lacked an inventive step. In subsequent letters, appellants 2 and 3 submitted further arguments.
- XI. In response to the written submissions of appellants 2 and 3, appellant 1 filed six sets of amended claims as auxiliary requests 2 to 7 and rebutted the objections raised.
- XII. Both appellants 2 and 3, respectively, made further written submissions in reply to the arguments presented by appellant 1.
- XIII. Oral proceedings were held on 28 October 2010.

During the course of the oral proceedings, appellant 1 submitted eleven sets of claims representing the main request and auxiliary requests 1 to 10.

The main request was identical to the previous main request, claim 1 thus having the wording quoted under point VI above.

Independent claim 1 of auxiliary request 1 has the following wording (amendments in respect of claim 1 of the main request highlighted by the board):

*"1. A method for treating fluoroaluminosilicate glass, comprising the steps of:*

a. *mixing finely-divided fluoroaluminosilicate glass with an acidic or basic aqueous silanol solution, **wherein the silanol is ethylenically-unsaturated,***  
and  
b. *drying the glass."*

Independent claim 1 of auxiliary request 2 has the following wording (amendments in respect of claim 1 of the main request highlighted by the board):

"1. A method for treating fluoroaluminosilicate glass, comprising the steps of:  
a. *mixing finely-divided fluoroaluminosilicate glass with an acidic or basic aqueous silanol solution **containing silanol in an amount of 0.1 to 20 % by weight, wherein the silanol is ethylenically-unsaturated,***  
and  
b. *drying the glass."*

Appellant 2 submitted at the oral proceedings an auxiliary procedural request called hereinafter "*handwritten request*".

The only objections regarding substantial issues raised by appellants 2 and 3 concerned lack of inventive step of the method according to claim 1 of the main request and the auxiliary requests submitted by appellant 1.

XIV. In addition to the documents mentioned under point II above, the following documents were referred to during the course of the appeal proceedings:



- O1D4a: Plueddemann, E.P.: *Silane Coupling Agents*;  
2nd edition, New York, London: Plenum Press,  
1991, p. 55 - 57;
- O2D6: EP 0 323 120 A;
- D13: *"3M F2000 Compomer Restorative System.  
Technical Product Profile"*. St. Paul, MN :  
3M, 1997, cover page and pages 3, 5 - 7, 12,  
13, 18 - 20.

Moreover, the parties referred to the following  
experimental evidence submitted during the course of  
opposition proceedings and the first appeal proceedings  
T 0184/00:

- E1: *"Experimental report"*, 15 pages, by Sumita B.  
Mitra, dated 4 October 1999 and filed by  
appellant 1 with letter of 8 October 1999;
- E2: *"Experimental report"*, 2 pages, by Sumita Mitra,  
submitted together with the grounds of appeal in  
case T 0184/00;
- D9: *"Testbericht"* by Almuth Bünsch, 3 pages, submitted  
by appellant 3 with letter dated 18 December 2000;
- D12: *"27.7.00 99262 Silanisierung nach 3M-Verfahren"*,  
excerpts from laboratory notes relating to the  
tests described in D9, 2 pages, submitted by  
appellant 3 with letter dated 21 October 2002;
- E3: *"Experiment A"* [and B], 1 page, submitted by  
appellant 1 with letter dated 6 December 2004;

E4: "*Comparative tests*", 4 pages, supplemented by an abstract (1 page) "*1823 Effect of Molecular Weight of Polyacid on Fluoride release from Glass-ionomer Cements*" by C. Shen et al., IADR, June 2003, submitted by appellant 2 with letter dated 14 January 2005;

E5: "*Versuchsbericht*" by Swen Neander, 5 pages, submitted by appellant 3 with letter dated 14 January 2005.

XV. As far as the arguments of the parties relate to the main request and the first and second auxiliary requests, they may be summarised as follows:

**Appellant 1** submitted that FAS glass cements, also known as "*glass ionomer cements*" were fluoride-releasing and, thus, cariostatic. A drawback of these materials was their mechanical fragility. According to D1, improved mechanical properties were obtained by treating the glass with polymers of  $\alpha, \beta$ -unsaturated carbonic acids. This led to a decrease of the fluoride release, however.

The present invention related to a method for treating FAS glass. The product thus obtained could be further treated with polyacids to produce a glass ionomer cement. For this purpose, any of the polyacids used in conventional glass ionomer cements, including light-curable liquids, were suitable. The treated FAS glass obtained according to the invention imparted on the glass ionomer cement good mechanical properties, but also an improved fluoride release. In this respect, appellant 1 referred particularly to example 1 and

comparative example 1 of the patent in suit, as well as to the experimental results contained in the reports E1, E2 and E3.

**Appellant 2** argued that the properties of the end product, i.e. the glass ionomer cements, were the result of the specific process conditions of their manufacture. Regarding these conditions, a number of choices had to be made, including the selection of the type of FAS glass to be used, the particle size, the type of silanes, acids or bases, as well as suitable process conditions in terms of temperature and processing time. None of these factors was specifically defined in claim 1 of the main request and the auxiliary requests.

Compared with the disclosure of D1, the claimed method was distinguished by a single feature, namely the use of an aqueous solution of a silanol. On the basis of the experimental evidence it was far from being established, however, that the treatment of the FAS glass with an aqueous solution of a silanol had a significant impact on the fluoride release of the final glass ionomer cements. Neither the examples contained in the description of the patent in suit, nor the results set out in the reports E1, E2 and E3 were conclusive in this respect. The alleged increase of the fluoride release of the glass ionomer cements made from FAS glass obtained according to the claimed method could be due to other factors than the treatment of the FAS glass. In particular, the results reported in E3 could be explained by the well known "*wash coat effect*".

Treating finely-divided FAS glass with a silanol containing solution followed by drying the glass was common in the dental field. Moreover, subjecting the FAS glass to an acidic environment was inherent in the preparation of a cement wherein acidic polymers were used. Fluoride release was an inherent property of FAS glass. Therefore, so appellant 2 argued, the proprietor of the patent in suit could not rely on these known effects. As far as the treatment with a basic aqueous silanol solution having a value of the pH from 8 to 12 was concerned, appellant 2 denied that there existed any effect at all.

Appellant 2 also submitted, that the experimental results contained in report E4 showed, that the presence of silanol in an amount of at least 0.1 wt% had no effect on the fluoride release properties of the glass in the presence of an acid. Any alleged effect *"represents an artefact and/or is expected by reasons known from the prior art."* Taking the results of report E5 into account, it even appeared that the highest fluoride release rate occurred in the absence of any silanation. This was in line with data contained in O2D6.

Therefore, taking D1 as the closest prior art, the technical problem was merely to find an alternative to the method for treating FAS glass disclosed in D1.

Regarding the incorporation of the feature of *"ethylenically-unsaturated"* silanol in claim 1 according to auxiliary requests 1 and 2, appellant 2 argued, that there was no evidence in support of the allegation, that the use of ethylenically-unsaturated

silanol gave rise to improved properties of the corresponding ionomer glass cements.

For all these reasons, appellant 2 concluded that the claimed method did not involve an inventive step.

Appellant 2 objected to the late filing of the auxiliary requests at the oral proceedings. Since the feature of "*ethylenically-unsaturated*" silanol was introduced into the respective claims 1 only in the course of oral proceedings, appellant 2 requested to be given an opportunity to provide further experimental evidence in case that the board considered one of auxiliary requests 1 to 9 to be allowable (see, in this respect, the "*handwritten auxiliary request*" quoted under point XVI below).

**Appellant 3** concurred with appellant 2, that the technical problem consisted merely in finding an alternative to the method for treating FAS glass disclosed in D1.

The product directly obtained by the claimed method was a treated FAS glass, not a glass ionomer cement ready to use. The latter included polymeric components in the form of a matrix formed by a curing process. None of the properties referred to by appellant 1 could be determined directly by means of tests performed on samples of the treated glass. Instead, samples of the final product, i.e. glass ionomer cements, had to be examined. The properties of glass ionomer cements depended to a large extent on the specific conditions used for converting the treated FAS glass into the final product. In this respect, the type of

polymerisable components used, as well as the conditions of the curing process were important factors. In order to achieve acceptable mechanical properties, both the "*cement liquid*" and the silanol coupling agent needed to contain polymerisable groups, in order to be capable of participating in the curing process. The claims did not, however, contain any features relating to this essential requirement.

Concerning the fluoride release, appellant 3 observed that glass ionomer cements made from untreated FAS glass provided the best results, whereas treatment of the glass with an aqueous silanol solution resulted in a decrease of the fluoride release. As to the impact of the use of an acidic aqueous silanol solution on the fluoride release, the experimental results contained in reports E2 and E3 did not demonstrate that the distinguishing features of the claimed method gave rise to an increased fluoride release rate. Variations of the fluoride release rate could be explained by other factors than the distinguishing features, including the mechanical treatment of the FAS glass and the drying conditions.

Referring to the report E5, appellant 3 denied that an increase of the fluoride release rate was obtained by the claimed method. But even assuming that some increase of the fluoride release rate occurred, this could only be regarded as a "*bonus effect*".

Appellant 3 observed that, in the absence of a proven technical effect in the form of an increased fluoride release, the technical problem, as defined by

appellant 1 on the basis of D1 taken as starting point, was not solved by the claimed method.

Furthermore, appellant 3 submitted, that the claimed method was obvious to the skilled person, having regard to D1 and the common general knowledge. D1 revealed, that the surface of FAS glass could be treated in "*conventional manner*" with an organic compound having an ethylenic double bond. This would have prompted the skilled person to convert silane into silanol before applying it to the glass, as suggested e.g. by O1D4a. In addition, the skilled person would have been aware of the need to operate under acidic or basic conditions, as suggested e.g. by documents O1D2, O1D3, O1D4, O1D5 and O3D4.

XVI. Requests

Appellant 1 requested, that the decision under appeal be set aside and that the patent be maintained on the basis of the claims according to the main request or according to one of the auxiliary requests 1 to 10, all requests filed during the oral proceedings.

Appellant 2 requested, that the decision under appeal be set aside, that the main request and the auxiliary requests 1 to 9 filed by appellant 1 during the oral proceedings be rejected and, in case that one of the auxiliary requests 1 to 9 was considered to be allowable, that the proceedings be continued in writing according to the handwritten auxiliary request filed by appellant 2 at the oral proceedings.

The *"handwritten auxiliary request"* referred to above has the following wording:

*"2. Alternatively, it is requested that the proceedings be continued in writing in order to give Opponent II an opportunity to provide experimental evidence showing that the feature "ethylenically-unsaturated" in relation to the term "silanol" cannot be used to establish "the maintenance of mechanical properties at an acceptable level" with regard to the closest prior art D1."*

Appellant 3 requested, that the decision under appeal be set aside, and that the patent be revoked in its entirety.

## **Reasons for the Decision**

1. *Admissibility of the requests submitted by appellant 1*
  - 1.1 At the oral proceedings, appellant 1 submitted in total 11 sets of claims representing the main request and auxiliary requests 1 to 10.
  - 1.2 The claims of the main request are identical to those submitted as main request as early as 21 November 2002, which request was admitted by the board and has been upheld throughout the proceedings.
  - 1.3 The wording of the claims of the first auxiliary request differs from the wording of the main request in that claim 1 is specified by including the feature *"wherein the silanol is ethylenically-unsaturated"*.



Moreover, claims 4 and 5 as granted were deleted and the remaining claims and back-references renumbered accordingly.

- 1.4 The wording of the claims of the second auxiliary request differs from the wording of the claims of the first auxiliary request in that claim 1 is further specified by the feature, according to which the aqueous silanol solution is "*containing silanol in an amount of 0.1 to 20 % by weight*".
- 1.5 The amendments effected to the claims of the first and second auxiliary requests were made in response to various arguments presented by appellants 2 and 3. The particular relevance of these arguments came into focus at the oral proceedings. The board is convinced, that said amendments are neither of particular complexity, nor do they raise any issues, that could not be expected to be dealt with by the parties without an adjournment of the oral proceedings.
- 1.6 Moreover, an auxiliary request relating to a set of claims specified by the feature of "*ethylenically-unsaturated*" silanol was submitted by appellant 1 already in the first appeal proceedings (see claim 1 of the first auxiliary request submitted at the oral proceedings in case T 0184/00), but a decision on this request did not become necessary in the earlier appeal and opposition proceedings. Therefore, the re-filing of such an auxiliary request cannot be considered as surprising.

1.7 The board also notes, that only appellant 2 raised an objection against the admissibility of the first and second auxiliary requests at the oral proceedings.

1.8 Having regard to the circumstances set out above, the board, in the exercise of its discretion conferred by Article 13(1) and (3) RPBA, admitted the main request and the first and second auxiliary requests of appellant 1 into the proceedings, in spite of their late filing.

1.9 Since the second auxiliary request can be granted (see below), there is no need to examine the admissibility of auxiliary requests 3 to 10 as submitted by appellant 1.

2. *Allowability of the amendments - main request and first and second auxiliary requests - Article 123(2) and (3) EPC*

2.1 The board is satisfied that the amendments effected by appellant 1 to the claims of the main request and the first and second auxiliary requests have a basis in the application as filed. Moreover said amendments serve to narrow down the scope of the concerned claims. This was not in dispute between the parties.

2.2 Thus, the amended claims are in conformity with the requirements of Articles 123(2) and (3) EPC.

*Main request*

3. *Claim 1 - Novelty*

3.1 Claim 1 of the main request is identical to claim 1 considered by the board in its previous decision T 0184/00. Novelty of the subject-matter of this claim was acknowledged by the board (see T 0184/00, p. 2 - 3, point II, second paragraph; p. 7 - 12, points 3 - 3.4).

3.2 The board is bound by the *ratio decidendi* of T 0184/00 with regard to the issue of novelty of claim 1 over the disclosure of D1. In T 0184/00 it is stated, that the method of claim 1 is novel over the disclosure of D1 at least in that the starting "aqueous" solution used to treat the FAS glass powder comprises a silanol (see decision T 0184/00, point 3.4 of the reasons).

3.3 The board is also satisfied that none of the other prior art documents relied upon by the appellants discloses a method with all the features of claim 1. This was not disputed.

3.4 Therefore, the subject-matter of claim 1 is novel (Articles 52(1) and 54(1)(2) EPC).

4. *Claim 1 - Inventive step*

4.1 The patent in suit relates to a method for treating finely-divided FAS glass with an acidic or basic silanol solution (see patent, page 2, lines 40 - 45). The FAS glass thus obtained can be formulated into fluoride-releasing and, therefore, cariostatic glass ionomer cements, which are useful in dentistry. For

producing such glass ionomer cements, the treated FAS glass can be combined *"in the presence of water with any of the polyacids used in conventional glass ionomer cements"* (see patent, page 2, lines 9 - 10; page 3, lines 7 - 8; page 6, lines 14 - 17).

4.2 It was common ground between the parties, and the board can accept this, that document D1 represents the closest prior art.

4.2.1 Document D1 belongs to the same technical field as the patent in suit and relates to dental ionomer cement compositions comprising an FAS glass powder. Like the patent in suit, document D1 addresses the problem of insufficient mechanical strength of glass ionomer cements. D1 teaches the use of a specific composition of the matrix material including, as a component "c", a polymerisable unsaturated organic compound containing an acryloyl or methacryloyl group (see D1, col. 1, line 44 to col. 2, line 23; claim 1).

4.2.2 Moreover, D1 describes methods for pre-treating FAS glass before processing it further into glass ionomer cements. According to a preferred embodiment, the glass powder is pre-treated with a polymerisable, ethylenically unsaturated, compound, for example an unsaturated carboxylic acid, or preferably a silane *"coupling agent"*, before it is incorporated into the cement mixture (see D1, claims 1 and 11; column 9, line 64 to column 10, line 35). For instance, in examples 6 and 14, a 10 % solution of  $\gamma$ -methacryloxypropyl-trimethoxysilane in ethanol was used in said pre-treatment, whereas in examples 7 and 15, vinyltriethoxysilane was used as coupling agent.

The finely-divided FAS glass was mixed with the ethanolic silane solution in a mortar and subsequently steam-dried at 110°C for 2 hours (see D1, column 14, lines 45 - 49; column 15, lines 24 - 28; column 19, lines 54 - 60; column 20, lines 41 - 45).

- 4.3 As regards the technical problem underlying the invention, appellant 1 submitted the following:
- 4.3.1 As confirmed in the patent in suit, a drawback of conventional glass ionomer cements consists in that they are relatively fragile. As a consequence, these products are prone to failure in high stress applications such as restoratives and crown core build-ups (see patent, page 2, lines 10 - 14; page 6, lines 28 - 30).
- 4.3.2 However, according to appellant 1, the methods disclosed in D1 gave rise to a new problem, namely low fluoride release rates of the glass ionomer cements obtained.
- 4.3.3 Accordingly, appellant 1 submitted at the oral proceedings, that having regard to the disclosure of D1, the technical problem was, thus, to be seen in providing a method for treating FAS glass, whereby cured glass ionomer cements, made from the treated FAS glass by mixing the finely-divided FAS glass powder with polyacids, exhibit an improved balance of properties, namely a high fluoride release rate and, at the same time, mechanical strength properties, which were acceptable for high-stress application (see also patent, page 2, lines 40 - 42; page 6, lines 18 - 19).

4.4 As the solution to said technical problem, the patent in suit proposes the method of claim 1 of the main request, which comprises the steps of:

- (a) mixing finely-divided FAS glass with an acidic or basic aqueous silanol solution, and
- (b) drying the glass.

4.5 The question arises, whether the technical problem as stated by appellant 1 is effectively solved by the claimed method, in particular, whether it is solved over the whole scope of claim 1. This was contested by appellants 2 and 3.

4.5.1 According to the patent in suit, the treated FAS glass can be made into a glass ionomer cement by combining it with polyacids that are capable of being cured, i.e. polyalkenoic acids such as a polyacrylic acid (see patent, page 6, lines 14 - 19).

4.5.2 At the oral proceedings the parties agreed, that the mechanical properties of glass ionomer cements depend essentially on the curing step, wherein the polymerisable matrix material, i.e. the polyacid, is a determinant factor. Moreover, there was a consensus, that crosslinking between the ethylenically unsaturated silanol on the one hand, which is used as the coupling agent of the FAS glass according to the patent in suit, and the polymerisable matrix material on the other hand, contributes significantly to the achievement of a higher mechanical strength of the cured glass ionomer cements.

Thus, it is not surprising, that silanol compounds comprising an ethylenically-unsaturated group lead to

higher values of the strength than silanol compounds containing no such polymerisable group. This is confirmed by the experimental data contained in the patent in suit, in particular by examples 2 to 22. Except in the case of examples 9 to 11, the FAS glass was treated with ethylenically-unsaturated silanols as coupling agents. In contrast, saturated silanols were used in examples 9 to 11. The cement liquids mixed with the treated FAS glass comprised ethylenically-unsaturated polycarboxylic acid copolymers (see patent, page 10, Table IV, "*Liquids A*" and "*Liquid B*"). It was found that the compressive strength (CS) and the diametral tensile strength (DTS) of the cement test samples according to examples 9 to 11 was generally lower than the strength of the test samples according to the examples with ethylenically-unsaturated silanols (see patent, pages 10 - 11, Table V, "*Ex No.*" 9 - 11, as opposed to "*Ex. No.*" 2 - 8 and 12 - 16; page 12, Table VII, "*Ex. No.*" 18 - 22).

4.5.3 However, it cannot be gathered from the available experimental data, that the replacement of the unsaturated silane "*coupling agent*" solutions exemplified in D1 by saturated silanols would lead to glass ionomer cements having simultaneously an acceptable strength and a higher fluoride release than glass ionomer cements obtained by the method of D1. On the contrary, the board concludes on the basis of the experimental results referred to above, that in order to obtain an acceptable level of mechanical strength, the silanols used for treating the FAS glass need to contain an ethylenically-unsaturated functional group, which allows crosslinking with the unsaturated polymerisable groups of the matrix material.

- 4.5.4 According to claim 1, any acidic or basic aqueous silanol solution may be used in the first step of the method. There is no limitation to ethylenically-unsaturated silanols that are capable of crosslinking with the unsaturated polymerisable groups of the matrix material. Consequently, for the reasons set out above, the technical problem is not credibly solved over the whole scope of claim 1.
- 4.6 Thus, the technical problem has to be reformulated in a less ambitious manner. In accordance with the submissions made by appellants 2 and 3 at the oral proceedings, the technical problem can merely be seen in providing a further, not necessarily improved method for treating FAS glass with a solution of a silanol coupling agent.
- 4.7 This less ambitious problem is evidently solved by the method according to instant claim 1.
- 4.8 It remains to be decided, whether or not the claimed solution was obvious in the light of the state of the art.
- 4.8.1 D1 discloses that ethylenically-unsaturated silanes may be applied as coupling agents *"in conventional manner"*, i.e. by mixing finely-divided FAS glass with a solution or suspension of the silane in a suitable solvent, followed by drying the glass (see D1, col. 10, lines 22 - 35). In the examples of D1, either ethanol (examples 6 - 7 and 14 - 16) or methanol (example 8) was used as the solvent.



4.8.2 The board holds, that the skilled person, when confronted with the technical problem stated above under point 4.6, would envisage using an aqueous solvent, the result being that at least a portion of the alkoxy-silane used in the method is hydrolysed to the corresponding silanol. In fact, it belongs to the common general knowledge of the skilled person, that organofunctional alkoxy-silanes may be hydrolysed before being applied to surfaces to function as coupling agents. Accordingly, it is a standard practice to use aqueous solutions of alkoxy-silanes as coupling agents to be applied to glass (see, for example, O1D4a, page 55, paragraph 3.1). Organotrialkoxy-silanes are commonly dissolved in water by shaking or stirring vigorously with acidified water until a clear solution results (see O1D4a, page 56, second paragraph). For these reasons, the skilled person would recognise without difficulty, that the use of an acidified solvent comprising water is, in fact, an obvious alternative to the use of methanol or ethanol as exemplified specifically in D1. Moreover, in view of the statement in D1, according to which FAS glass may be treated with silane coupling agents "*in conventional manner*", the skilled person would expect comparable mechanical properties when applying the silane coupling agents in form of acidified aqueous solutions comprising at least some silanol.

4.8.3 When following the above approach, the skilled person would arrive without the exercise of inventive skills at a method for treating FAS glass falling within the ambit of claim 1. Hence, claim 1 embraces subject-matter not involving an inventive step (Article 52(1) and 56 EPC).

5. For these reasons the main request cannot be allowed.

*First auxiliary request*

6. *Claim 1 - Novelty*

6.1 Compared with claim 1 of the main request, claim 1 of the auxiliary request 1 is further specified by the feature, that the **silanol is ethylenically-unsaturated**.

6.2 From the fact, that the present claim 1 is narrower in scope than claim 1 according to the main request, it follows, that its subject-matter is also novel (Article 52(1) and 54(1) (2) EPC).

7. *Claim 1 - Inventive step*

7.1 With regard to the first auxiliary request, appellant 1 maintained, that in view of the closest prior art represented by D1, the technical problem was the one stated under point 4.3.3 above.

7.2 As the solution to the technical problem, a method for treating FAS glass according to claim 1 of the first auxiliary request is now proposed. Said method comprises the following steps (emphasis added by the board):

- (a) mixing finely-divided FAS glass with an acidic or basic aqueous silanol solution, **wherein the silanol is ethylenically-unsaturated**, and
- (b) drying the glass.

7.3 Again, appellants 2 and 3 contested, that the technical problem as formulated by appellant 1 was solved over

the whole scope of claim 1, *inter alia* because it had not been demonstrated, that the desired results were achieved, irrespective of the amount of silanol in the acidic or basic aqueous silanol solution. Since neither a lower, nor an upper limit of the amount of silanol was indicated in claim 1, the claim was too broad in scope.

- 7.3.1 At the oral proceedings, appellant 1 did not deny, that under certain conditions the desired high values of the fluoride release rate or a particularly high level of the mechanical strength were not obtained. This did not mean, however, that the problem was not solved over the whole scope of claim 1. When carrying out the claimed method, the skilled person would naturally use his common general knowledge in order to select suitable conditions and avoid any mistakes. It could be expected from the skilled person to have a sound knowledge of the normal working practice and the routine means in the technical field of glass ionomer cements.
- 7.3.2 The board is not convinced by the argument of appellant 1, according to which there is no need to define at least the lower limit of the amount of silanol to be used.
- 7.3.3 In this respect, appellant 1 referred to the experimental results contained in report E3, according to which there was a marked effect in terms of an increase of the fluoride release rate, even if the amount of hydrolysed silanol was as low as 0.1 % by weight. Appellant 1 agreed, however, that the technical problem was not solved in the complete absence of any

amount of hydrolysed silanol. But such an embodiment of the method did not correspond to the method according to claim 1.

7.3.4 According to E3, a silanol solution containing 0.1 % by weight of hydrolysed silanol gave rise to a fluoride release rate of 56.6 ppm after 3 days and 98.5 ppm after 5 days. In contrast, values of 38.3 ppm after 3 days and 52.6 ppm after 5 days were measured in the case of the treatment solution according to example 6 of D1 (see E3, page 1, experiments A and B).

7.3.5 At the oral proceedings, appellants 2 and 3 maintained, that the unit "ppm" used in E3 for characterising the fluoride release was unusual, unclear and, thus, meaningless. In response, appellant 1 explained, that in all experiments the same measurement method and units were used as in Table VI of the patent in suit (see patent, page 11, lines 37 - 54). The unit of "ppm" was equivalent to the unit of " $\mu\text{g/g}$ ".

7.3.6 The board notes, that the unit of "ppm" is not only used in E3 submitted by appellant 1, but also in the reports D9 and E5 submitted by appellant 3 (see D9, page 3, drawing "*Fluorid Freisetzung*"; E5, page 4, Table; page 5, Abb. 3). This indicates, that the unit of "ppm" is not as unusual in the relevant technical field, as pretended by appellant 2. Indeed, E3 contains neither an explicit definition of the unit "ppm", nor a description of the method used for determining the fluoride release of the test samples. In spite of this, the board accepts, that the results reported in E3 demonstrate, that a significant relative increase of the fluoride release can be achieved using a solution

according to the claimed method and containing 0.1 % by weight of silanol, as opposed to the use of a solution according to example 6 of D1a (see E3, page 1, last paragraph, "*Fluoride release in ppm*").

- 7.3.7 The further argument of appellant 2, according to which the results could be explained by a "*well known wash coat*" effect was rejected by appellant 1.

The post-published abstract annexed to E4 mentions an "*initial surface washcoat*". However, in the absence of any evidence in support of the occurrence of a "*wash coat*" effect and its impact on the experimental results contained in E3, the board regards the argument of appellant 2 merely as an unsubstantiated allegation.

- 7.3.8 Thus, the board is satisfied, that an amount of 0.1 % by weight of hydrolysed silanol is sufficient to solve the technical problem as formulated by appellant 1 (see above, point 4.3.3).

- 7.3.9 This does not mean, however, that significantly lower amounts of hydrolysed silanol still produce the desired effects in terms of the fluoride release rate and the mechanical properties. The acknowledgment of the alleged improvement would require, that said effects be achieved to a degree distinguishing them from the effects obtained by the method of the closest prior art D1. In this respect, the board notes, that according to the patent in suit, the silanes are converted into the corresponding silanols only during the treatment under the influence of water, the pH of which is adjusted with acid or base to provide a non-neutral solution (see patent, page 2, lines 43 - 45; pages 9 - 16,

examples 1 - 42). This implies, that whenever no significant hydrolysis (depending on time) has occurred, the amount of silanol in the aqueous silanol solution is negligible. At low levels of hydrolysis practically the whole amount of the coupling agent contained in the treatment solution is present in the form of silane, as in the method of D1 (see, for example, D1, column 14, lines 45 - 48). It is only reasonable to assume, that under such conditions the effects achieved cannot be distinguished from the ones obtained when using the method of D1.

7.3.10 For these reasons the board arrives at the conclusion, that in the absence of any indication in claim 1 of a minimum level for the amount of hydrolysed silanol, the technical problem is not solved over the whole scope of the claim.

7.4 Therefore, as in the case of the main request, the technical problem has to be reformulated. It can again merely be seen in providing a further method for treating FAS glass with a silane solution disclosed in D1.

7.5 Said technical problem is evidently solved by the method of the present claim 1.

7.6 However, having regard to the fact, that the use of ethylenically-unsaturated silanes is expressly disclosed in D1 (see D1, column 10, lines 10 - 18), the additional feature of claim 1 of the first auxiliary request does not render the claimed method inventive.

7.7 Thus, said claim 1 also encompasses methods which do not involve an inventive step (Article 52(1) and 56 EPC).

7.8 For these reasons the first auxiliary request is not allowable either.

*Second auxiliary request*

8. *Claim 1 - Novelty*

8.1 Compared with claim 1 of the first auxiliary request, claim 1 of the second auxiliary request is further specified by the feature, according to which the silanol solution **contains silanol in an amount of 0.1 to 20 % by weight.**

8.2 From the fact, that claim 1 is narrower in scope than claim 1 according to the main request, it follows that its subject-matter is also novel (Article 52(1) and 54(1) (2) EPC).

9. *Claim 1 Inventive step*

9.1 Regarding the second auxiliary request, appellant 1 also maintained, and the board accepts it, that the technical problem to be solved in the light of the closest prior art D1 was the one stated under point 4.3.3 above.

9.2 As the solution to said technical problem, the patent now proposes a method for treating FAS glass comprising the following steps (emphasis added by the board):

- (a) mixing finely-divided FAS glass with an acidic or basic aqueous silanol solution **containing silanol in an amount of 0.1 to 20 % by weight**, wherein the silanol is ethylenically-unsaturated, and
- (b) drying the glass.

9.3 Appellants 2 and 3 held that said technical problem was not effectively solved by the claimed method over the whole scope of claim 1.

9.3.1 In view of the evidence relied upon by appellant 1, the board is, however, satisfied, that the technical problem is, in fact, successfully solved by the claimed method over the whole scope of claim 1.

(i) Example 1 and comparative example 1 of the patent in suit illustrate the effects of the claimed method, compared with the method according to the closest prior art D1. In example 1, untreated FAS glass powder was mixed with an aqueous solution of "A-174" (i.e.  $\gamma$ -methacryloxypropyltrimethoxysilane, see O1D5, page 1, right hand column, lines 1 - 3 and formula). The mixture was acidified to pH 3.5, using trifluoroacetic acid (TFA), and stirred for one hour, followed by further stirring for 4.25 hours and drying overnight in a 45 °C oven (see patent, page 9, lines 14 - 20).

Comparative example 1 was carried out in accordance with examples 6 and 7 of both documents D1 and D1a, with some minor variations (see patent, page 8, lines 46 - 51; D1, column 14, lines 45 - 49; col. 15, lines 25 - 28; D1a, col. 13, lines 26 - 30; col. 14, lines 1 - 4). It was found that, using the same cement liquid, the cured glass ionomer cement of example 1



exhibited a much greater cumulative fluoride release than the cement of comparative example 1, for example 362.0 µg/g after 12 days, as opposed to 51.8 µg/g in the case of comparative example 1 (see patent, page 9, lines 31 - 43, Table III). Regarding the mechanical properties, the glass ionomer cement of example 1 showed an average compressive strength (CS) of 152 MPa and an average diametral tensile strength (DTS) of 29 MPa, compared with an average CS of 130 MPa and an average DTS of 14.3 MPa in comparative example 1 (see patent, page 9, lines 24 - 28; page 9, lines 5 - 8).

Apart from these comparative data, further evidence for the effects of the method according to claim 1 is provided by a number of examples performed under both acidic and basic conditions (see patent, pages 9 - 17, examples 2 to 8, 12 to 23, 25, 26 and 28 to 42).

(ii) In a further series of experiments reported in E1, appellant 1 compared the compressive strength (CS) and the diametral tensile strength (DTS) of samples of glass ionomer cements obtained from FAS glass treated with various solutions of "A-174" containing (i) ethanol and water, (ii) absolute ethanol, or (iii) methanol, water and acetic acid. The treated glass powder was dried in a tray at 110 °C in an oven for 2 hours (see E1, page 2, line 21 to page 3, line 16, experiments 2A, 2B and 2C). In experiment 2C, the FAS glass was treated under the same conditions as in example 25 of the patent in suit, whereas experiments 2A and 2B reproduced the conditions of example 6 of D1 and D1a. Significantly higher values for CS and DTS were obtained in experiment 2C, compared with the

values obtained in experiments 2A and 2B (see E1, page 3, lines 17 - 30, Table 1).

(iii) Appellant 1 submitted another report E2, concerning the treatment of FAS glass with:

- an aqueous, alcohol containing solution of silane "A-174" in the absence of an acid or base (experiment 1A);
- a neutral aqueous, alcohol containing solution of silanol originating from "A-174" (experiment 1B); and
- an acidic aqueous, alcohol containing solution of silanol originating from "A-174" in the presence of acetic acid (experiment 1C). The treated glass was dried in a plastic tray at 110 °C for 6 hours (see E2, page 1, experiments 1A, 1B, 1C). Subsequently samples of glass ionomer cements were prepared, using as cement liquid the "liquid B" according to the patent in suit (see patent, page 10, lines 28 - 38, Table IV). Under neutral conditions using a silane solution (experiment 1A), 130 MPa were found for CS, 20 MPa for DTS and 28.6 "g/g" (*recte* µg/g, see patent, page 9, line 34) after one day and 76.2 µg/g after 7 days for the cumulative fluoride release. Under the conditions of experiment 1B, using a neutral aqueous silanol solution, increased values of 148 MPa for CS, 24 MPa for DTS and 40.2 µg/g after one day and 112.4 µg/g after 7 days for the fluoride release were observed. Still higher values were found under acidic conditions (experiment 1C), namely 156 MPa for CS, 30 MPa for DTS and 110.4 µg/g after one day and 308.0 µg/g after 7 days the cumulative fluoride release. (see E2, page 2, lines 5 - 10, results).

(iv) Appellants 2 and 3 criticised, that the experimental evidence referred to by appellant 1 related to the properties of glass ionomer cements made from treated FAS glass rather than to those of the FAS glass itself. Yet, the conversion of the treated FAS glass into the final glass ionomer cements did not form part of the features of claim 1. In any case, neither the examples contained in the patent in suit, nor the reports E1, E2 and E3 were conclusive. Various factors not mentioned in claim 1 could be responsible for the results obtained in the experiments. In this context, appellants 2 and 3 mentioned not only the selection of "A-174" as the coupling agent, but also the type and particle size of the FAS glass, the specific acid or base used for adjusting the pH of the treatment solution, the processing time, the temperature, the drying conditions, the form of curing, as well as the period of time, after which the fluoride release was determined.

(v) The board accepts, that the factors mentioned by appellants 2 and 3 may have an impact on the properties of glass ionomer cements. Nevertheless, in practice the skilled person will have to determine suitable process conditions, when carrying out the claimed method. Thereby the skilled person will rely on common general knowledge. The need to apply reasonable process conditions when putting the method into practice does not necessarily mean, that the scope of claim 1 is too broad. In other words claim 1 cannot be regarded as being too broad in scope for the mere reason, that the distinguishing features are not exclusively responsible for the fluoride release rate and the mechanical strength of the final glass ionomer cements. In the

present case, what needs to be established is, whether the distinguishing features make a significant contribution to the result to be achieved, i.e. a relative improvement of the properties of the cured glass ionomer cements.

The board is satisfied, that in the present case the experimental evidence submitted by appellant 1, in particular example 1 and comparative example 1, demonstrate, that such a contribution is entailed by the distinguishing features of claim 1.

9.3.2 For the board, the arguments and counter evidence of appellants 2 and 3 are not conclusive for the following reasons:

(i) As far as the reports E1 and E2 are concerned, appellant 3 contested the results. He submitted an experimental report D9 complemented by excerpts from laboratory notes D12 as counter-evidence, with a view to proving:

- that untreated FAS glass gave rise to the highest fluoride release;
- that, irrespective of whether FAS glass is treated with neutral or acidic aqueous solutions of "A-174", the fluoride release rates were the same within the error margin; and
- that the values for compressive strength (CS) of glass ionomer cements made from untreated FAS glass und from FAS glass treated with "A-174" were equal (see D9, page 2, Table 1; page 3, Figure).

The board is not convinced by the results reported in D9, complemented by D12, because in the experiments of

D9 the test samples were compomers, not resin-modified glass ionomer cements as in the patent in suit and D1. Referring to D13, appellant 3 argued, that this difference was not of importance, because the transition between conventional glass ionomer cements, resin-modified glass ionomer cements and compomers was only gradual. The board notes, however, that according to D13 compomers have in general a lower fluoride release than glass ionomer cements, although in certain cases the fluoride release may be similar (see D13, page 6, first paragraph, lines 10 - 12; page 18, last paragraph, lines 1 - 5; page 19, Figure 8). Therefore, the experiments of D9 do not disprove the results presented in the patent and the reports E1 and E2 (see also decision T 0184/00, point 4 of the reasons, page 14, line 29 to page 15, line 2).

(ii) Incidentally, appellant 2 observed at the oral proceedings, that claim 1 excluded only the value of exactly 7 for the pH of the aqueous silanol solution, all other values being either acidic or basic. Appellant 2 questioned, whether treatment of FAS glass with a silanol solution having a pH very close to 7 resulted in a higher fluoride release rate than in the case of the method of D1.

The board considers, that this argument does not hold water. In the patent in suit, it is stated, that the treatment solution is adjusted with an acid or base to provide a non-neutral solution in order to promote the hydrolysis of the silane employed to produce a sufficient amount of silanol (see patent, page 2, lines 43 - 45). The results of E3 show, that a silanol solution in a mixture of water and absolute alcohol

having a weight ratio of 4 : 1 and adjusted with ammonium hydroxide to a pH of 6.9 was effective (see E3, page 1, second paragraph, lines 2 - 6; last paragraph, "*Exp.A*"). It goes without saying, that the skilled person will only contemplate values of the pH, which are sufficiently remote from exactly neutral conditions (i.e. a value of the pH equal to 7) to give rise to the formation of the required amount of silanol by hydrolysis of the corresponding silane component. In practice, the skilled person does not need any inventive skills to determine suitable values of the pH in a specific case.

(iii) Furthermore, appellant 2 submitted a report E4 containing experimental data, including non-cumulative fluoride release data in the form of the fluoride release per week of a glass ionomer cement produced under nearly the same conditions as the product of example 1 of the patent in suit (experiment 1). For comparative purposes, the fluoride release rate of an identical glass ionomer cement, except that silane was omitted, was determined (experiment 2). It was found that within experimental error, the fluoride release rates from the third to the eighth weeks were the same, irrespective of whether the treating solution contained silane, or not (see E4, page 2, Table).

Furthermore, appellant 3 referred at the oral proceedings also to examples 10 and "*Control A*", as well as examples 24, 27, 31 and 32 of the patent in suit (see patent, page 13, Table X, "*Run No. 3*" and "*Control E*"; page 15, Table XI). Both "*Control E*" and example 27 related to untreated glass. In the view of appellant 3, these examples showed, that the mechanical

properties of glass ionomer cements obtained from FAS glass treated according to the claimed method (examples 10, 24, 31, 32) were not consistently better than the properties glass ionomer cements obtained from untreated FAS glass ("Control A", "Control E", example 27). Thus, the mechanical properties (CS, DTS and  $K_{IC}$ ) of examples 31 and 32 were worse than those of "example 27", which was in reality a comparative example.

On the basis of E4 and the examples referred to above, appellants 2 and 3 concluded that, generally speaking, glass ionomer cements made from untreated glass exhibited higher fluoride rates than cements made from glass treated according to the claimed method. Moreover, the mechanical properties of glass ionomer cements based on untreated FAS glass were better. If, in the case of treated FAS glass, acceptable values for compressive strength (CS), diametral tensile strength (DTS) and fracture toughness ( $K_{IC}$ ) were achieved, the reason was not the treatment of the FAS glass, but the choice of the matrix material. Therefore, the claimed method did not solve the technical problem.

The board is not convinced by this argumentation for the following reasons:

E4 relates to a comparison of the fluoride release of a glass ionomer cement made from FAS glass treated according to the claimed method, as opposed to the fluoride release of a glass ionomer cement based on untreated FAS glass. Such a comparison is not meaningful, however, because the reference sample, i.e. the cement made from untreated FAS glass, does not

represent the closest prior art D1. For this reason alone, the results reported in E4 are not conclusive.

As to examples 10 and 24 of the patent in suit, they are not relevant either, because they relate to silanes having no ethylenically-unsaturated groups, namely "T2924" (N-trimethoxysilylpropyltributylammonium bromide) and "A-1100" ( $\gamma$ -aminopropyltriethoxysilane) (see patent, page 11, line 10; page 13, line 10; page 3, lines 24 - 25, 27). The use of such saturated silanes is not encompassed by the present claim 1.

There remains only the comparison between the examples 31 and 32 on the one hand, and example 27 on the other hand. Again, this comparison is not conclusive. Firstly, because example 27 relates to untreated FAS glass, i.e. not to the closest prior art (see above), and secondly because different polyacids were used for making the respective glass ionomer cements. In example 27, polyethylene glycol dimethacrylate with an average of 4.5 ethyleneglycol units ("PEG<sub>200</sub>DMA") was mixed with the treated FAS glass, whereas in the case of example 31 the average of ethyleneglycol units was 9 ("PEG<sub>400</sub>DMA"), and in the case of example 32 the average was 13.6 ("PEG<sub>600</sub>DMA") (see patent, page 15, Table XI, "Ex." 27, 31, 32; page 4, lines 48 - 50). Justifiably, appellants 2 and 3 emphasised at the oral proceedings, that the choice of the matrix material was important.

(iv) In a still further report E5, appellant 3 submitted experimental data regarding the fluoride release rate, compressive strength (CS) and diametral tensile strength (DTS) of samples of glass ionomer cements made from untreated FAS glass and from FAS



glass treated under neutral and acidic conditions. Two different cement liquids having a composition similar to ("*in Anlehnung an*"), but not identical with the cement liquid of example 6 of D1 were used. Since the samples obtained from the first cement liquid could not be properly cured, all measurements were performed with samples based on the second cement liquid. In the experiments reported in E5, the highest cumulative fluoride release rate was obtained with untreated FAS glass, whereas treatment under neutral conditions resulted in decreased rates, which were further reduced under acidic conditions (see E5, page 5, Abb. 3). Regarding the diametral tensile strength (DTS), the lowest value of CST were obtained with untreated FAS glass, whereas treatment under neutral and acidic conditions led to improved values (see E5, page 3, Abb. 1). According to appellant 3, the experiments showed that the choice of the cement liquid was decisive regarding the fluoride release rate. Depending on the specific cement liquid used, the fluoride release was either increased as in E2, or reduced as in E5. In the view of appellant 3, therefore, E5 proved that treatment of FAS glass with an aqueous silanol solution according to the claimed method does not lead invariably to glass ionomer cements having increased fluoride release rates.

The board is not convinced by this argumentation. Regarding the mechanical properties of glass ionomer cements reported in E5, the results appear to be inconsistent with those of E2 (see above). The board accepts, however, that the discrepancies result from the fact, that different cement liquids were used for preparing the test samples in the experiments of E2 and

E5. According to E5, the cement liquid contained an unspecified "*partially esterified polyacrylic acid*", glycerol-DMA and a component called "*HEMA*" (see E5, page 2, lines 10 - 16, Table). In contrast, the tests of E2 were performed with "*Liquid B*" having the composition specified in Table IV of the patent in suit (see patent, page 10, lines 27 - 39, Table IV, column to the right). Given the differences of the cement liquids used, appellant 1 questioned at the oral proceedings, whether the tests of E5 were carried out in a skilful manner. The board has no reason to assume that they were not. However, for the board, a single comparative test between a glass ionomer cement based on FAS glass treated according to the claimed method, and a reference cement produced in analogy to the method of D1, but with a different cement liquid, cannot be regarded as evidence of sufficient strength to disprove the experimental results contained in the patent in suit.

(v) Having regard to the reasons set out above, neither E5 nor the other experimental reports submitted by appellants 2 and 3 convince the board, that the technical problem underlying the invention (see above, point 4.3.3) is not solved over the whole scope of claim 1. In this respect, appellants 2 and 3 have not discharged the burden of proof resting on them.

9.4 Since the stated technical problem (see above, point 4.3.3) is credibly solved over the whole scope of claim 1, it remains to be decided, whether or not the claimed solution was obvious to the skilled person.

9.4.1 The closest prior art D1 is concerned with the physical properties of the glass ionomer cements, in particular with the deterioration of the physical properties of the hardened mass under the influence of moisture (see D1, column 1, lines 44-50). In order to obtain glass ionomer cements exhibiting a low sensitivity to water at the initial stage of setting, D1 proposes a composition comprising *inter alia* (a) a polymer of an  $\alpha,\beta$  unsaturated carboxylic acid having an average molecular weight of 5'000 to 40'000; (b) an FAS glass powder capable of reacting with said polymer; and (c) a polymerisable unsaturated organic compound having at least one  $\text{CH}_2=\text{C}(\text{R}_1)-\text{COO}-$  group, wherein R1 is H or  $\text{CH}_3$  (see D1, column 3, lines 18-27; claim 1). The FAS glass may be treated on its surface in known manners with an organic compound having a polymerisable, ethylenically unsaturated double bond, most preferably a silane coupling agent (see D1, column 9, line 64 to column 10, line 35).

9.4.2 The method for treating FAS glass disclosed in D1 solves the technical problem of insufficient physical properties, in particular in terms of compressive strength, under the influence of water at the initial stage of curing (see D1, col. 1, line 36 - 50; col. 9, line 64 - col. 10, line 10; and e.g. examples 6 and 7).

9.4.3 Yet, in the present case the technical problem was not simply to achieve a high compressive strength at the initial stage of curing. What was required was a good balance between the fluoride release rate and the mechanical strength, expressed by parameters like compressive strength (CS), diametral tensile strength

(DTS) and fracture toughness  $K_{IC}$ , of glass ionomer cements comprising treated FAS glass.

9.4.4 D1 is not specifically concerned with the fluoride release rate of the cement, let alone with the proper balance between the fluoride release and the mechanical strength properties. On the basis of the comparative example 1 of the patent in suit, it can be concluded, that the method according to D1 leads to glass ionomer cements having rather unbalanced properties, namely good values for compressive strength (CS) and diametral tensile strength (DTS), but a low fluoride release rate (see patent, page 9, lines 5-8, 24 - 27; Table III, column "*comparative example 1*"). Therefore, D1 taken alone does not provide any guidance as to how the technical problem (see above, point 4.3.3) can be solved.

9.4.5 Therefore, it has to be investigated, whether the other documents belonging to the state of the art contain a teaching, which prompts the skilled person to modify the method for treating FAS glass according to D1 in a manner leading to a method having all features of present claim 1.

9.4.6 In this respect, appellants 2 and 3 emphasised, that the skilled person knew from D1, that FAS glass could be treated "*in known manner*". What was meant by this, could be derived from the data sheet of "A-174", i.e. O1D5, where it was stated that "A-174" was soluble in various organic solvents, but also "*in water when suitable hydrolysis procedures are followed*" (see O1D5, page 2, section "*solubility*"). Moreover, the skilled person knew from O2D3, O1D4 and O1D4a, that silanol

groups were required for the organofunctional silanes to function as coupling agents between organic resins and substrates, such as silicate glass or mineral fillers (see O2D3, page 34, right hand column, lines 5 - 8 and 31 - 33; O1D4, page 33, lines 6 - 7; O1D4a, page 55, section 3.1, lines 1 - 5). Another relevant disclosure was the statement in O3D4, according to which any organic silane of the formula  $R-Si(R^1)(R^2)(R^3)$  was suitable for the treatment of silica fillers, provided that at least one of the groups R,  $R^1$ ,  $R^2$  and  $R^3$  was a hydroxy group or a group that can be hydrolysed into a hydroxy group, which means that the concerned compounds have the chemical structure of silanols (see O3D4, page 3, last paragraph, line 29 to page 4, first paragraph). Appellants 2 and 3 argued, that these documents provided to the skilled person the information, which was needed to arrive at the claimed method.

- 9.4.7 The board is not convinced by this argumentation. None of the documents O1D4, O1D4a, O1D5, O2D3 or O3D4 relates to the treatment of FAS glass or the properties of glass ionomer cements.

Documents O1D4, O1D4a and O2D3 are extracts from textbooks dealing with the use of silane coupling agents in general. None of the documents addresses the problem of finding a proper balance of the fluoride release rate and the mechanical strength of glass ionomer cements.

Document O1D5 is a product brochure from the manufacturer of the organofunctional silane "A-174". It discloses the use of "A-174" as primer for clay or

glazed tiles, as coupling agent for glass roving size-binder formulations, as additive in filled polyester resin composites, and as additive in filled thermoplastic resin systems or finish on glass in reinforced resin systems (see O1D5, page 2, sections 1, 2 and 3; page 3, section 4). The issues of the fluoride release rates and the mechanical properties of glass ionomer cements are not addressed in the document.

Document O3D4 relates to dental composites containing a polymerisable component and a silanised filler, such as silica glass powder (see O3D4, claim 1; page 4, second and third paragraphs; page 8, example A). Again, neither the fluoride release, nor the mechanical properties of glass ionomer cements are addressed in the document.

Therefore, the skilled person would not consider these documents in the expectation of finding a solution to the technical problem underlying the invention.

- 9.4.8 The same applies to documents O1D2, O1D3, O2D2 and O3D5, which were also referred to by appellants 2 and 3.

O1D2 describes a dental filling material comprising fused silica treated with a basic aqueous solution of vinyl silane (see O1D2, column 6, claim).

Document O1D3 is concerned with a process for preparing stable solutions of silanols, comprising the step of hydrolysing a silane in an aqueous acidic solution, followed by extracting the silanols with an organic solvent. The organic solution thus obtained may be used for treating mineral dental filling materials or glass

(see O1D3, col. 1, claims 1 and 2; col. 3, lines 29 - 33; col. 4, lines 19 - 26). FAS glass is not mentioned in the document. Accordingly, the issue of the fluoride release rate is not addressed.

Document O2D2 relates to resin-based fluoride releasing composites containing FAS glass, which is preferably silanised (see O2D2, claim 1; page 7, line 9; page 11, examples 7 and 8; page 11, line 51 "*silanated WP*"; page 14, line 17 "*silanated XT glass*"). The document does not provide any information regarding the method used for silanating the glass.

Document O3D5 discloses dental composites containing an FAS glass filler and a resin component. It is stated in the document, that the filler should be is treated with a silane coupling agent, preferably  $\gamma$ -methacryloxypropyltrimethoxysilane (see O3D5, page 1, first paragraph; page 3, last paragraph; page 4, third paragraph). However, there is no disclosure in O3D5 of a method for said treatment with a silane coupling agent.

9.4.9 The board observes, that the good values for the fluoride release obtained in accordance with the claimed method cannot be regarded as a mere "*bonus effect*" as alleged by appellant 3. In view of the plurality of factors potentially having an impact on the fluoride release, and in view of the many conceivable manners for treating FAS glass, the effects of the claimed method are neither accidental, nor the result of a "*one way street*" situation. Thus, basic requirements for establishing a "*bonus effect*" are not met.

9.4.10 The board observes further, that the synopsis of D1 with each of the documents invoked by appellants 2 and 3 fails to suggest a method having all features of the method according to claim 1. In fact, nowhere in the documents referred to above, there is any disclosure, that the acidic or basic aqueous silanol solution to be used for the treatment of FAS glass has to contain silanol in an amount of 0.1 to 20 % by weight.

9.4.11 Under these circumstances, the board arrives at the conclusion that the skilled person was not induced by the state of the art to modify the method disclosed in D1 in a manner leading to a method falling within the terms of claim 1.

9.5 Having regard to the foregoing, the board is satisfied, that the method for treating FAS glass according to claim 1 of the second auxiliary request was not obvious to the skilled person. Thus, the method of claim 1 involves an inventive step, as required by Articles 52(1) EPC and 56 EPC.

10. *Dependent claims 2 to 23*

Dependent claims 2 to 23 all depend directly or indirectly on independent claim 1. Consequently, the subject-matter of these claims is also novel and inventive (Articles 52(1) 54(1) (2) and 56 EPC).

11. *Product claims 24 to 26*

11.1 Independent product claim 24 and claims 25 and 26 dependent thereon have the same wording as claims 35



to 37 of the patent as granted, except for their numbering and their references to higher-ranking claims. The claims are directed to specific polymeric alkoxysilanes.

11.2 Throughout the opposition proceedings and the first appeal proceedings T 0184/00, no objections were raised against claims 35 to 37 as granted, let alone were such objections duly substantiated under the grounds of opposition invoked (Article 100(a) EPC). This was expressly addressed in the decision under appeal (see reasons, point 3, second paragraph). Nevertheless, although appellants 2 and 3 requested the revocation of the patent in its entirety, they did not submit in the present appeal proceedings, be it in writing or during the oral proceedings, any facts, evidence or arguments in support of an objection under Article 100(a) EPC against said product claims.

11.3 Hence, as far as instant claims 24 to 26 are concerned, the issues of novelty and inventive step do not fall within the factual framework of the appeal proceedings (Article 12(1), (2) RPBA). Under these circumstances, the board sees no reason for considering said issues in the present appeal proceedings (see also decision T 0223/05 of 24 April 2007, points 2.1 and 2.2 of the reasons).

11.4 Consequently, the second auxiliary request submitted by appellant 1 is allowable.

*Third to tenth auxiliary requests*

12. In view of the board's finding concerning the second auxiliary request, there is no need to deal with the remaining third to tenth auxiliary requests.

The *"handwritten auxiliary request"* of appellant 2

13. At the oral proceedings, appellant 2 questioned, whether the limitation to *"ethylenically-unsaturated"* silanols could *"establish the maintenance of mechanical properties at an acceptable level with regard to the closest prior art"*.

13.1 However, appellant 2 did not present a specific argumentation in support of this query. Instead, appellant 2 requested, that the appeal proceedings be continued in writing, in order to give him the opportunity to provide experimental evidence in support of his allegation.

13.2 However, the board observes, that the question raised by appellant 2 appears to be at odds with the latter's earlier acknowledgment concerning the contribution of vinyl groups contained in silanols to the mechanical stability of glass ionomer cements, whenever used in combination with unsaturated carboxylic acids as cement component.

13.3 Moreover, the feature in question was present in dependent claims 4 and 5 of the patent as granted. In addition, nearly all examples contained in the patent in suit relate to silanol having one or more vinyl groups. It was therefore recognisable without any difficulty, that using ethylenically-unsaturated silanols was a particularly preferred feature of the

method according to the patent in suit. Also, as already mentioned, a request to incorporate in claim 1 the feature of "*ethylenically-unsaturated*" silanol was already submitted in the first appeal proceedings T 0184/00 (see above).

13.4 Therefore, the board holds, that appellant 2 could and should have raised this issue much earlier than at the oral proceedings. Under these circumstances, it would be inappropriate to prolong the present appeal procedure by giving appellant 2 a further opportunity to submit experimental evidence of unknown type, still to be performed and hence open as to the results it would provide.

13.5 For these reasons, the board decided not to allow the "*handwritten auxiliary request*" of appellant 2.

**Order**

**For these reasons it is decided that:**

1. The decision under appeal is set aside.
2. The case is remitted to the department of the first instance with the order to maintain the patent on the basis of the claims 1 to 26 according to the second auxiliary request filed by appellant 1 during the oral proceedings, and a description to be adapted.
3. The handwritten request of the appellant 2 filed during the oral proceedings is rejected.

The Registrar:

The Chairman:

C. Vodz

B. Czech