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**D E C I S I O N**  
of 16 December 2003

**Case Number:** T 0511/01 - 3.2.3  
**Application Number:** 93115191.4  
**Publication Number:** 0590477  
**IPC:** E04C 2/02, E04C 1/40  
**Language of the proceedings:** EN

**Title of invention:**

Architectural material using metal oxide exhibiting photocatalytic activity

**Patentee:**

TAKENAKA CORPORATION

**Opponents:**

SAINT-GOBAIN VITRAGE  
RHODIA CHIMIE

**Headword:**

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**Relevant legal provisions:**

EPC Art. 52, 54, 56, 123(2)

**Keyword:**

"Main request (not new)"  
"First auxiliary request (added subject-matter)"  
"Second auxiliary request (inventive step, yes)"

**Decisions cited:**

T 0651/91, T 0508/91

**Catchword:**

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Case Number: T 0511/01 - 3.2.3

**D E C I S I O N**  
**of the Technical Board of Appeal 3.2.3**  
**of 16 December 2003**

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**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 7 March 2001  
revoking European patent No. 0590477 pursuant  
to Article 102(1) EPC.

**Composition of the Board:**

**Chairman:** C. T. Wilson  
**Members:** J. du Pouget de Nadaillac  
J. P. B. Seitz

## Summary of Facts and Submissions

I. The appeal is directed against the decision posted on 7 March 2001 of an Opposition division of the European Patent Office, which revoked the European patent EP-B-0 590 477 for lack of novelty or inventive step of the subject-matter of amended claims of said patent. The patentee - hereinafter the appellant - filed the notice of appeal on 28 April 2001 and paid the appeal fee on the same day. Together with the statement of grounds of appeal, which was received on 7 July 2001, he submitted two sets of claims as main and auxiliary request. The claims of these two sets correspond respectively to the claims of the main and fifth auxiliary request, which were rejected in the impugned decision.

II. Claim 1 according to main request reads as follows:

"An architectural material comprising:

- a base (16) of quartz glass or tile having a light-receiving surface and used as an architectural material as an external wall material, a roofing material, an internal wall material, a flooring material or a ceiling material; and
- a titanium oxide thin film (14) exhibiting photocatalytic activity formed by coating a surface of said quartz glass or tile having the light-receiving surface with a titanium oxide sol, optionally with titanium dioxide powder added thereto, and by sintering the same."

III. Opponents O1 and O2, hereinafter the respondents, by letters received on 10 and 7 January 2002 respectively, merely referred to their submissions filed before the first instance

IV. In response to the board's communication pursuant to Article 11(2) RPBA in which the board expressed its provisional opinion of the case with respect to novelty and inventive step, the appellant filed on 10 May 2003 a new set of claims 1 to 12 for the auxiliary request.

Claim 1 of this set has the following wording:

"An architectural material comprising

- a base (16) of quartz glass or metal tile having a light-receiving surface and used as an architectural material as an external wall material, roofing material, an internal wall material, a flooring material or a ceiling material; and
- a titanium oxide thin film (14) having deodorising and antimold properties and imparting these properties to the architectural material, and exhibiting photocatalytic activity formed by coating a surface of said quartz glass or tile having the light-receiving surface with a titanium oxide sol with titanium dioxide powder added thereto, and by sintering the same."

V. Oral proceedings took place on 17 June 2003. Respondent O2 had informed the board by fax sent on 21 February 2003 that they would not attend these proceedings and,

accordingly, they were not present or represented. In accordance with Rule 71(2) EPC, the proceedings were continued without them.

During these proceedings, the appellant submitted two new sets of claims as second and third auxiliary requests.

VI. The two independent claims, namely claims 1 and 5, according to the second auxiliary request have respectively the following wording:

"An architectural material including a surface and its vicinity which are substantially formed of a metal mixture including a metal oxide (50),

characterized in that

- said architectural material (16) includes an external wall material, a roof material, an internal wall material, a flooring material or a ceiling material,
- said metal oxide exhibits photocatalytic activity, and
- said metal mixture includes a second metal (54) for improving the photocatalytic activity of said metal oxide,
- said architectural material (16) includes an inner portion which is substantially formed of a metal mixture including a metal (52) of the same kind as that which constitutes said metal oxide and said second metal for improving the photocatalytic activity of said metal oxide, and said surface and



D1: Translation of JP-220677/92,  
first priority document of D.

D5: FR-A-2 600 550

D6: "Electrochemical synthesis and in-situ Raman  
spectroscopy of thin films of titanium dioxide" by  
Arsow, Korman and Plieth, Journal of Raman  
Spectroscopy (1991), Vol.22, pages 573 to 575.

F: EP-A-0 306 301

G: JP-61-083106 (G2: English translation)

H: English abstract of JP-01-218635 (WPI)

M: US-A-4 773 717

IX. The appellant defended the patent in suit as follows:

Both documents C and G disclose an hydrolysis step before the calcinating or firing step, so that it cannot be said that the conditions for the coated product to be fired are the same as those of the coated product to be sintered according to claim 1 of the main request. The heating step according to both documents serves to remove crystal water from the coating and, thus, is not a sintering step.

Between the disclosure of document D and that of its priority document D1, at least two differences can be seen, namely the porous aspect of the titanium oxide film and the anatase type of the titanium oxide, so that the first priority date is not valid and, as a

consequence, document D is not a prior art citation under Article 54(3) EPC.

Thus, claim 1 of the main request is new and implies an inventive step.

In the Embodiment I of the description of the patent in suit, as originally filed, the term "tile" covers any kind of tile, and, thus, the introduction of the expression "metal tile" in claim 1 of the first auxiliary request does not infringe Article 123(2) EPC. The argumentation of the respondent concerning claim 1 of the second auxiliary request supposes a person skilled in the art having a great imagination. Moreover, document M concerns a semi-transparent reflective glass, which has no catalytic activity, and document H does not concern an architectural material. Therefore, the skilled person has no reason to combine the teaching of one of these documents with that of the citation C1.

X. Respondent 01 essentially argued as follows:

The subject-matter of claim 1 according to the main request is not new, having regard to either C1 or D1: C1 discloses for example a tile having on its surface a semiconductor such as titanium dioxide exhibiting a photocatalytic function, which is used to prevent pollution. According to pages 4 and 7 of this prior art, the metal oxide is coated on the substrat surface and then the coated substrat is calcined at from 350 to 700° C during one hour in an oxidising gas atmosphere. In D1, a decorative material is made of a support made of glass, metal and the like, on which a film of



titanium dioxide is applied by a coating method, and then said decorative material is fired at temperatures between 300 to 800°C for two hours.

The subject-matter of claim 1 according to the second auxiliary request, which corresponds to claim 3 of the main request, is also not new in view of the teaching of C1, since this document - see the last lines of page 4 or claim 2 - also teaches the use of a second metal such as platinum to be carried by the titanium dioxide layer. The person skilled in the art receiving this information will mix both metals, namely titanium and platinum, and oxidise the mixture, reaching thereby a structure of the architectural material surface and its vicinity in accordance with the wording of the contested claim 3.

Supposing that this claim is considered as new, it nevertheless does not imply an inventive step, combining the teaching of either document C1, F or G with that of document M. Documents C1, F or G (see claim 2 of this last document) teach the skilled person that the addition of a metal such as platinum or palladium improves the photocatalytic activity of titanium dioxide or the like. Thus, the skilled person knows that he has to bring this metal in the titanium oxide film and he will look in the technical field of coating to see how he can do so, while improving simultaneously the peeling resistance of the film. Document M gives him the answer by disclosing a method of forming a thin film of a metal on a support and oxidising said film on its surface.

XI. The appellant requested that the decision under appeal be set aside and that the patent be maintained on the basis of either:

- (1) his main request (claims 1 to 12) filed on 7 July 2001 together with the statement of grounds of appeal,
- (2) his first auxiliary request (claims 1 to 12) filed on 10 May 2003,
- (3) his second auxiliary request (claims 1 to 10) filed during the oral proceedings,
- (4) his third auxiliary request (claims 1 to 9) filed during the oral proceedings.

The respondent requested the appeal to be dismissed.

### **Reasons for the decision**

1. The appeal is admissible.

#### *Main request*

2. In the description of the patent in suit, the sintering step mentioned in the last feature of claim 1 was one of several cited possibilities for preparing the metal oxide film and is described solely in connection with the examples of Embodiment I, in which temperatures of 400° or 200°C for 30 minutes are given. The heat treatment according to the last paragraph of column 4 can - in the case of titanium oxide - be effected in

the temperature range of from 100 to 800°C for about 20 to 120 minutes in an electrical furnace. This heat treatment is said to improve the film strength and the bonding force with respect to the base. The patent in suit provides no other information as to the sintering step.

Thus, in the patent in suit, the expressions "sintered" and "heat treated" have the same meaning and include temperatures between 100 to 800°C applied from 20 to 120 minutes on an architectural basic material having a surface already coated with a metal oxide sol, for example a titanium dioxide sol.

Thus, according to the content of the patent, only the times and temperatures of sintering are to be considered. If in a prior art document the same corresponding parameters are disclosed in combination with the same starting product, it has to be assumed that a sintering process occurs, even if the term "sintering" or "sintered" has not been used. One cannot give as sole reason for the novelty criterion a "special treatment" without having disclosed all the conditions which are necessary to perform this treatment, unless it can be proven that for a person skilled in the art before the priority date of the patent in suit other conditions would be assumed automatically to apply. During the oral proceedings, the appellant did not bring forward such conditions.

3. For the use of oxide metals having photocatalytic function, such as titanium dioxide, which are coated on a base surface to prevent pollution of said surface, document D1 discloses many applications, for example

fountains, ponds, aquarium walls and tiles, so that architectural materials are concerned. On page 4, several possible known coating methods are given, for example a dipping method in which the base to be surface treated is immersed in a solution containing a titanium compound, pulled up and dried and then calcinated. A hydrolysing step may occur before the calcining step, but such a step is only given as being optional. Since the calcination occurs at from 350 to 700°C in an electrical furnace for about 1 hour - see pages 4 and 7 of D1 -, it corresponds to the heat treatment or "sintering" according to the patent in suit. Therefore, no difference can be seen between the architectural material described by this prior citation and that according to claim 1 of the main request.

Thus, the subject-matter of this claim 1 is not new and, as a consequence, the main request is not allowable (Articles 52 and 54 EPC).

*First auxiliary request*

4. In claim 1 of this request, it is claimed that the base can be made of metal tile. However, as mentioned above, the sintering step in the patent in suit concerns only the Embodiment 1 and, according to the documents as originally disclosed of this patent, see the first lines of page 7, glass and tiles are disclosed in this embodiment as architectural materials, however not metal tiles. Metal tiles are only mentioned in relation with Embodiment II, which does not include a sintering step.

The argument of the appellant that the term "tile" supports all kinds of tiles, including metal tiles, cannot be followed. A generic disclosure does not constitute a disclosure of the specific examples falling within this generic disclosure (Guidelines C-IV, 7.4, confirmed by the decisions T 651/91 and T 508/91).

Thus, there is no support in the originally filed documents of the patent in suit for the specific example "metal tile", as architectural material which has to be subjected to a sintering treatment. Hence, added subject-matter has been introduced in claim 1 of the first auxiliary request, which therefore is not allowable, infringing Article 123(2) EPC.

*Second auxiliary request*

5. The features of the two independent claims of this request, namely claims 1 and 5, are based on the disclosure of Embodiment II in the original description of the patent in suit and respectively on claims 6 and 10 as originally filed, which correspond to the granted claims 5 and 8.

The description and drawings have been adapted to these new claims, all the passages relating to the first embodiment in the description as originally filed being deleted. Figures 1 to 7 correspond to the Figures 15 to 21, as originally filed.

Thus, the requirements of Article 123(2) and (3) EPC are met.

6. Document C1 in its claim 2 and on page 4, indeed, discloses that a platinum metal may be carried by or loaded on the titanium oxide layer. However, this prior art document does not indicate that the platinum metal is mixed with the titanium oxide. In contrast thereto, it is disclosed in the first lines of page 5 that the chloroplatinic acid is attached to the metal oxide layer and then subjected to decomposition. In the following disclosed Example 1 - see page 7 of C1 - the architectural material, after being coated with the titanium oxide film and calcinated, is immersed in a chloroplatinic acid/ethanol solution, pulled up and dried, and "then thermally decomposed at 200°C to obtain a quartz glass plate sample having on the surface thereof a titanium oxide layer carrying platinum". No idea of a mixture is given. Therefore, contrary to the respondent's opinion, document C1 does not disclose all the features of claims 1 and 5.

Document F apparently does not concern an architectural material of the kind concerned by the subject-matter of claims 1 and 5 (see the following paragraph 9 of the present decision). Moreover, it describes neither a surface of an architectural material comprising the progressive mixture of metals and metal oxide(s) according to claim 1 of the above request nor a method implying the oxidation of a mixture of metals.

Document D constitutes prior art under Article 54 (3) EPC, however only in respect of its first priority date; since the first priority document D1 does not disclose the use of added metals which could improve the photocatalytic activity of the titanium oxide film, document D is not relevant for the invention as here claimed.

Among the other above cited documents, which may concern architectural material, documents A1, B1 and G2 teach as C1 the use of a second metal, such as platinum and palladium, for improving the photocatalytic action of a semiconductor metal oxide coated on a base surface. However, they all teach to deposit this second metal on the metal oxide thin film (page 3 of A1; pages 2 and 9 of B1; pages 4, 5 and 7 of G2). A1 and G2 even give precise details of the methods of depositing the second metal on the already formed metal oxide film, as was the case with C1, the various known coating methods and moreover photodecomposition being mentioned. In none of these documents is a mixture of both metals, as first step, mentioned.

Thus, the subject-matter of both claims 1 and 5 is new (Articles 52 and 54 EPC).

7. The architectural material according to claim 1 of the second auxiliary request is obtained by the method of claim 5. In the description of the patent in suit, it is disclosed that the solution according to these claims 1 and 5 improves the strength and peel resistance of the architectural material.
8. For respondent O1, the starting point for the invention here claimed can be either C1 or F or G2; in the impugned decision, A1 was considered for this purpose.
9. The selection of document F for representing the closest prior art is not logical, since this prior art does not clearly concern an architectural material having " a surface and its vicinity" of the kind

according to the first characterising feature of claim 1: In F, a mesh substrate in the form of a sleeve made of several layers of fiberglass strands is shown for the base, since it is wished to have a construction having coated surfaces for the titanium oxide as large as possible, while simultaneously being porous, that is to say with holes, so that it has to be transparent to light to ensure that all coated surfaces receive such light and the fluid to be purified can pass through it. Moreover, a single sentence on page 3, lines 56 and 57, indicates that "enhanced results can sometimes be achieved by doping the active material (that is to say the metal oxide film) with a suitable dopant, e.g. platinum." How this doping is realised is not disclosed. Only coating methods for the metal oxide film such as those disclosed in A1 or C1 are described.

It may be that the term "doping" in document F suggests to apply a mixture of the titanium oxide and the second metal on the base. However, there is no suggestion in this document either to apply said mixture in a way such that the progressive structure according to claim 1 should be obtained or to subject the base, once coated with this mixture, to an oxidation process (so that the claimed structure should be obtained). Such an oxidation process should be at variance with the teaching of F, page 3, line 36, to use titanium alkoxide as starting element.

10. G2 provides the same following teaching as C1: the second metal is added after the formation of the titanium oxide film on the base and is applied by the immersion or dipping technique followed by a heat treatment.



11. It is already not clear for which reason a person skilled in the art would wish to depart from the above given methods of applying the second metal, especially as in these documents heat treatments are mentioned, which are known to provide good bonding forces between the coated surface and the coating film, as is the case for the solution of the present invention according to the main request. Moreover, document A1, page 3, teaches that, when the titanium oxide film is irradiated by light, electrons and holes are generated inside the film and then move to the surface of the film, react with the moisture adhering to the surface to be oxidised and ultimately turned to CO<sub>2</sub>. It is also explained that the presence of the second metal supported on the TiO<sub>2</sub> film will improve the efficiency of the said photocatalytic action. Thus, the skilled person is advised that the essential effect occurs on the surface of the thin film part and is consequently not led to introduce the second metal into the inner portion of the titanium oxide film, in which apparently it would have no effect.

Thus, the disclosures of these documents C1 and G2 do not suggest to mix the second metal with the titanium oxide.

12. Document M concerns a different technical field, describing methods of making glass articles with decorative colour coatings, which can comprise metal oxide films. The aim of this prior art is to provide a semi-transparent reflecting article. There is no clear link between this technical field and that of the present invention, so that the citation of this

document is to be seen as the result of an *a posteriori* search.

Furthermore, the application of a metal film on a substrate with subsequent oxidation is one of many methods, which are disclosed in this citation for applying a dielectric thin film layer, and the reason for selecting this particular method is not grounded. In particular, there is no indication or suggestion that this method increases the peel resistance of the coated base. It also does not concern a mixture of two metals or of a metal with a metal oxide, so that it cannot suggest to apply the above mentioned particular coating method comprising the oxidation step to a mixture of the metals or metals oxides, which are disclosed in documents A1, G2 or even F.

13. Summarising, having regard to the documents cited by the respondent 01, the subject-matter of claims 1 and 5 implies an inventive step (Articles 52 and 56 EPC).
14. In his submissions before the first instance, respondent 02 objected to the lack of inventive step of the subject-matter of claims 1 and 5, having regard either to a combination of document B1 with D5, or D6 alone.
  - 14.1 As mentioned above, B1 takes into account that platinum or palladium increases the photocatalytic activity of titania (the kind of titanium oxide, which exhibits this action).

However, platinum is said to be expensive, so that the solution proposed in B1 is to bring the photocatalytic

material such as titania on a support which is a conductive material, so that a photoelectrochemical effect is obtained. Any kind of conductive metal can be used and in Example 1 of B1, which was mentioned by respondent 02, titania is flame-coated on a stainless steel plate. This teaching does not correspond to the subject-matter of claim 1, which requires the second metal to be a metal "for improving the photocatalytic activity of the metal oxide". As known in the state of art and as acknowledged in the description of the patent in suit, under this expression, particular metals such as platinum, rhodium or palladium, and not any kind of conductive metals, are meant. In fact, B1, by suggesting to replace these particular metals by a conductive metal, goes away from the present invention, as claimed in claims 1 and 5. Under these circumstances, it is not necessary to examine the combination of B1 with D5. Moreover, this last document, as recognised by the respondent, does not concern the coating of a mixture of metals.

- 14.2 D6 is a study on the crystal modifications occurring into a thin titanium dioxide film during the electrosynthesis of titanium oxides on titanium surfaces, the titanium oxides being obtained by electrochemical oxidation of titanium electrodes. It is briefly indicated that titanium oxide has many uses, e.g. as photocatalyst. According to the respondent, by oxidizing titanium electrode, the product obtained has a surface made of oxide metal, and then progresses inwardly to an inner part made of pure metal.

No hint is given in this citation to apply titanium oxide to an architectural material, so that already it

is not clear how this document could lead the skilled person to the subject-matter of claims 1 and 5. There is also no mention in this prior art of a second metal, which would improve the photocatalytic action of the titanium oxide, so that the preliminary conditions to think about a mixture of metals are lacking.

## 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 an amended form with the following documents:
  - Claims 1 to 10 according to the second auxiliary request filed during the oral proceedings;
  - Description: pages 2 to 4 and 8 to 10, as filed on 21 August 2003, and pages 5 and 11, as filed on 24 September 2003;
  - Drawings, as filed on 21 August 2003.

The Registrar:

The Chairman:

D. Sauter

C. T. Wilson