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**Datasheet for the decision  
of 15 May 2012**

**Case Number:** T 0431/09 - 3.3.07

**Application Number:** 95101593.2

**Publication Number:** 666107

**IPC:** B01J 23/74, A62D 3/00,  
B01J 37/26, B01J 37/02,  
B01J 21/06

**Language of the proceedings:** EN

**Title of invention:**  
Titanium oxide photocatalyst

**Patent Proprietors:**  
ISHIHARA SANGYO KAISHA, LTD.

**Opponents:**  
KRONOS INTERNATIONAL, INC.

**Headword:**  
-

**Relevant legal provisions:**  
EPC Art. 100b), 83, 111(1)

**Keyword:**  
"Sufficiency (yes)"  
"Remittal for further prosecution"

**Decisions cited:**  
-

**Catchword:**  
-



Case Number: T 0431/09 - 3.3.07

**D E C I S I O N**  
of the Technical Board of Appeal 3.3.07  
of 15 May 2012

**Appellants:** ISHIHARA SANGYO KAISHA, LTD.  
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**Representative:** Henkel, Breuer & Partner  
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**Respondents:** KRONOS INTERNATIONAL, INC.  
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**Decision under appeal:** Decision of the Opposition Division of the  
European Patent Office posted 12 December 2008  
revoking European patent No. 666107 pursuant to  
Article 101(3)(b) EPC.

**Composition of the Board:**

**Chairman:** J. Riolo  
**Members:** F. Rousseau  
M.-B. Tardo-Dino

## Summary of Facts and Submissions

- I. The appeal by the Patent Proprietors (Appellants) lies from the decision of the Opposition Division posted on 12 December 2008 revoking European patent No. EP-B-0 666 107 granted on the basis of European patent application No. 95 101 593.2.
- II. A notice of opposition had been filed requesting revocation of the patent in its entirety on the grounds of lack of novelty and inventive step (Article 100(a) EPC) and lack of sufficiency of disclosure (Article 100(b) EPC). The documents submitted before the Opposition Division included the following:
- D1 Palmisano et al., The Journal of Physical Chemistry, Vol. 92, No. 23, pages 6710-6713, 1988,
  - D2 Cordischi et al., Journal of Solid State Chemistry, Vol. 56, pages 182-190, 1985
  - D3 Photocatalysis, Fundamentals and Applications, edited by N. Serpone and E. Pelizzeti, John Wiley & Sons, pages 159 and 168-173, 1989, and
  - D4 Bockelmann et al., Solar thermal utilization, Vol. 6, pages 397-429, 1992.
- III. The decision was based on claims 1 to 5 submitted during the oral proceedings on 26 November 2008 as the Patent Proprietors' sole request, claim 1 reading as follows:
- "1. A titanium oxide photocatalyst for use in synthesis of organic substances or decomposition

of noxious materials under irradiation of ultraviolet rays, comprising titanium oxide particles having an iron compound in an amount of 0.0005 - 10% by weight in terms of Fe based on the weight of TiO<sub>2</sub> of the titanium oxide particles, which are obtained by hydrolysis or neutralization of a titanium compound in an aqueous solution containing the iron compound dissolved therein, and subsequent treatment with a mineral acid, said titanium oxide particles having an average particle size of 1 to 50 nm, and having said iron compound supported in an amount of 0.05 to 5,000 µg in terms of Fe per square meter of the surface of the titanium oxide particles."

- IV. According to the contested decision, amended claim 1 fulfilled the requirements of Articles 123(2) and (3) and 84 EPC, as well as those of Rule 80 EPC. Having regard to sufficiency of disclosure, it was held that the range of 0.05 to 5000 µg Fe per square metre related only to the amount of iron supported on the surface, i.e. using the Opposition Division's own wording "*on the top*" of the photocatalyst particles. In order to carry out the claimed invention, the skilled person would not only have to produce the particles using example 2 as his main guidance, but would also have to determine the Fe content on the surface of the photocatalyst particles to determine whether or not the results fell within the scope of claim 1. Since the iron concentration at the particle surface strongly influenced the photocatalytic activity, this feature had to be considered as essential and had therefore to be determined accurately. In the absence of any suitable definition of the surface of the photocatalyst,

and of any guidance on how to determine the concentration of iron on the surface the photocatalyst particles, the skilled person had to rely on his general knowledge. The methods proposed by the Patent Proprietors for measuring the iron concentration on the surface of the particles, i.e. secondary ion mass spectrometry as a physical method and chemical leaching of the surface of the photocatalyst particles, was however not suitable for quantitatively determining iron on the surface of particles having a size of 1 to 50 nm. The Opposition Division was not aware of any other method such as AES, XPS, XRF or ICP-AES which would enable the skilled person to measure the concentration of Fe on the surface of the photocatalyst particles having a size as small as 1 nm and concentration of iron as low as 5 ppm. The fact that in example 2 (the only remaining example according to the invention) the iron concentration on the surface seemed to have been calculated as the ratio of the total Fe content and the specific surface area did not show that such a method had to be employed, as it would rely on the assumption that the iron was always completely present on the surface of the particles, which assumption was not correct in view of the synthesis employed for producing the photocatalyst particles. Hence, the skilled person would have to develop a new method or adapt a known method for measuring the iron concentration. This did not only constitute an undue burden, but also necessitated an inventive step. Consequently, the claimed subject-matter and the invention to which it related were not disclosed in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art.

- V. With the statement setting out the grounds of appeal dated 22 April 2009, the Appellants argued that the skilled person was aware that the most appropriate method to quantitatively determine the iron concentration on the surface of the claimed titanium oxide particles comprised in a first step chemical leaching of the titanium oxide particles using hydrochloric acid at a concentration of 5-10%, in a second step precipitation of the dissolved iron with cupferron and in a third step calcination of the precipitate, which allowed an amount of  $\text{Fe}_2\text{O}_3$  to be determined.
- VI. In response thereto, the Respondents submitted in a letter of 29 October 2009 that the method indicated by the Appellants for measuring the amount of iron on the surface of the titanium oxide particles, first, was not indicated in the patent in suit and, second, as shown by an experimental report submitted with the same letter, did not provide reproducible results.
- VII. With a letter dated 2 November 2011, the Appellants submitted a set of claims 1 to 5 as Auxiliary Request and additional arguments supporting their contention that the skilled person would be in a position to reliably determine the amount of iron supported on the surface of titanium oxide particles, using in a first step, chemical leaching.
- VIII. The parties were summoned to attend oral proceedings to take place on 15 May 2012. In a communication dated 28 December 2011 sent in preparation of the oral proceedings, the Board gave a reasoned preliminary opinion that no case had been made for lack of

sufficiency. Attention was drawn to the Opponents' written submissions of 4 June 2007 and to exhibit A1 submitted with their letter of 10 August 2008. The Board indicated in essence that the wording "said iron compound supported in an amount of 0.05 to 5,000 µg in terms of Fe per square meter of the surface of the titanium oxide particles" was to be understood in the light of the examples of the patent in suit as the ratio of the amount of iron of the particles (expressed in wt.-% based on the weight of the TiO<sub>2</sub>) to the specific surface area of said particles.

- IX. The Appellants requested that the decision under appeal be set aside and that the patent be maintained on the basis of the set of claims underlying the contested decision, i.e. submitted on 26 November 2008 during the oral proceedings before the Opposition Division (Main Request), or alternatively on the basis of claims 1 to 5 submitted with letter of 2 November 2011 (Auxiliary Request). It was also requested that the case be remitted to the Opposition Division for further prosecution.
- X. The Respondents requested that the appeal be dismissed.
- XI. At the end of the oral proceedings, the decision of the Board was announced.

## Reasons for the Decision

1. The appeal is admissible.

### *Main Request*

2. Amendments made to the claims in the course of opposition appeal proceedings are to be fully examined as to their compatibility with the requirements of the EPC, in particular with regard to the provisions of Article 123(2) and (3) EPC (G 9/91, OJ 1993, 408, point 19 of the reasons). However, apart from the replacement of the expression "having an iron compound supported" by "having said iron compound supported", the subject-matter of claim 1 of the present Main Request corresponds to that of claim 2 as granted. Under these circumstances the Board was not entitled to examine the clarity of present claim 1, as any ambiguity concerning the meaning of this claim in respect of the features "*subsequent treatment with a mineral acid*" and "*having said iron compound supported in an amount of 0.05 to 5,000 µg in terms of Fe per square meter of the surface of the titanium oxide particles*" (see below) was already present in the claims as granted. In the absence of any ground of opposition raised by the Opponents under Article 100(c) EPC, the Board also did not address and *a fortiori* did not take a decision on the issue of whether or not the combination of features defined by claim 1 of the Main Request, which defines the same object as claim 2 as granted, extends beyond the content of the application as filed. It also follows from the above that claim 1 of the Main Request, as it corresponds in essence to claim 2 as granted, is in keeping with the requirements of Article 123(3) EPC.



3. The issue to be decided in the present appeal proceedings is whether or not the Opposition Division was right to find that the patent in suit did not disclose the claimed invention in a manner sufficiently clear and complete for it to be carried out by a person skilled in the art. It is the established jurisprudence of the Boards of Appeal that the requirements of sufficiency of disclosure are only met if the invention as defined in the claims can be performed by a person skilled in the art in the whole area claimed without undue burden, using common general knowledge and having regard to further information given in the patent in suit. The invention as defined in claim 1 of the Main Request relates to a titanium oxide photocatalyst for use in synthesis of organic substances or decomposition of noxious materials under irradiation of ultraviolet rays, which comprises titanium oxide particles containing an iron compound, said particles being obtained:

- (i) by hydrolysis or neutralisation of a titanium compound in an aqueous solution containing the iron compound dissolved therein, and
- (ii) subsequent treatment with a mineral acid,
- (iii) said particles having an average particle size of 1 to 50 nm and an amount of iron compound of 0.0005 - 10% by weight in terms of Fe based on the weight of  $\text{TiO}_2$  of the titanium oxide particles, and

(iv) having said iron compound supported in an amount of 0.05 to 5 000 µg in terms of Fe per square metre of the surface of said titanium oxide particles.

4. It is not in dispute that step (i), i.e. hydrolysis or neutralisation of a titanium compound in an aqueous solution containing an iron compound dissolved therein, is a well known technique for preparing titanium oxide photocatalysts comprising an iron compound, as illustrated by D1 (page 6711, "catalyst preparation"), D2 (page 183, point 2.1 "Materials") and (D4 (page 404, point 3). Details concerning this technique, including suitable starting compounds, are given in paragraphs [0013] and [0014] of the patent in suit. The patent in suit also provides in paragraphs [0034] and [0038], under the headings "Example 1" and "Example 2" respectively, examples of synthesis of titanium oxide particles comprising an iron compound through hydrolysis of a titanium compound in an aqueous solution containing an iron compound dissolved therein.
5. Concerning feature (ii), i.e. a subsequent treatment with a mineral acid, claim 1 does not indicate the purpose of that process step, nor is this purpose implicit from claim 1, which does not contain any specific information concerning that step. Moreover, none of the passages of the specification which relate to methods comprising hydrolysis or neutralisation of a titanium compound in an aqueous solution containing an iron compound dissolved therein refers explicitly to a "subsequent treatment with a mineral acid". In the passage of the patent labelled "Example 2" (paragraph [0038]), which does not use the wording

"treatment with a mineral acid", nitric acid is merely used, in line with the teaching in paragraph [0014] mentioning the use of a "monobasic acid such as nitric acid or hydrochloric acid", to adjust the pH to 1.5 for achieving peptisation of the product resulting from hydrolysis of titanyl sulfate. The passages of the patent in suit which explicitly refer to a treatment with a mineral acid, namely paragraphs [0004], [0016] and [0017], relate to a treatment of titanium oxide particles which are not disclosed to contain iron, the mineral acid being indicated to provide some dissolution of the titanium oxide. As already implicitly indicated above, it is not mentioned that the treatment with a mineral acid is carried out subsequently to step (i) as defined in present claim 1. Although the specification, as follows from the above, does not provide any guidance for a treatment with a mineral acid after having performed step (i), the Board considers that the skilled person who is not requested by present claim 1 to achieve any particular effect when treating the product resulting from step (i) could nevertheless carry out process step (ii) merely by contacting the product resulting from claim 1 with a mineral acid.

6. Summing up, the mere definition in claim 1 of a treatment with a mineral acid subsequent to step (i), despite the lack of information or restriction to defines the function this treatment serves, which issue might nevertheless be considered when assessing inventive step, does not lead *per se* to a lack of sufficiency of disclosure.

7. It is furthermore not disputed that the skilled person by applying steps (i) and (ii) and on the basis of his general knowledge regarding preparation of titanium oxide particles comprising iron compounds by hydrolysis or neutralisation of a suitable starting compound, is able to obtain particles which exhibit an average particle size of 1 to 50 nm and contain an amount of iron compound of 0.0005 - 10% by weight in terms of Fe based on the weight of TiO<sub>2</sub> of the titanium oxide particles.
  
8. Thus, it remains to be examined whether the skilled person is able without undue burden, using common general knowledge and having regard to further information given in the patent in suit, to provide the above particles with "*iron compound supported in an amount of 0.05 to 5,000 µg in terms of Fe per square meter of the surface of said titanium oxide particles*". The Respondents argue that owing to the absence of any mention with respect to the measurement method for determining amounts of iron supported on the surface of the titanium oxide nanoparticles according to claim 1, the skilled person would not be able to quantify the amount of iron bound on the surface of the particles and as a consequence would not be able to distinguish that amount from the amount of iron present within the particles (i.e. in the volume defined by the particles). It was in particular pointed out that no reliable method existed for assessing such a parameter for particles having a size between 1 to 50 nm.
  
9. The first issue to consider in this respect is the meaning of the expression "*iron compound supported in an amount of 0.05 to 5,000 µg in terms of Fe per square*

*meter of the surface of said titanium oxide particles".* Neither claim 1 as granted, in which this expression can be found, nor the specification defines or provides an explicit basis for understanding this expression in the same manner as the Respondents in the appeal proceedings, namely as the amount of iron which is supported on the surface of the particles, as opposed to the amount of iron contained within the particles. In the absence in the claims or in the general part of the description of the patent in suit of any explicit definition for this amount of iron expressed in  $\mu\text{g}$  iron per square metre of the surface of said titanium oxide particles, the skilled person would turn to the experimental part of the specification, which relates to the preparation of samples A to D, F, G, I, K, N and O, for which amounts of iron compound supported per square metre of the surface of the titanium hydroxide particles are indicated  $\mu\text{g}/\text{m}^2$ . The skilled person inevitably would note, like the Opponents did in their notice of opposition of 4 June 2007 and with their letter of 10 August 2008 on the basis of exhibit A1 submitted therewith, that the phrase "*amount of iron hydroxide (resp. iron compound) supported per square meter of the titanium oxide particles*" used for the embodiments exemplified in the patent in suit expresses an amount indicated in  $\mu\text{g}/\text{m}^2$ , which merely corresponds to the ratio of the amount of iron (expressed in wt.-% based on the weight of the  $\text{TiO}_2$ ) to the specific surface area of the  $\text{TiO}_2$  particles (given in  $\text{m}^2/\text{g}$ ). In addition to the fact that this finding is valid for any of the samples of titanium dioxide particles comprising iron disclosed in the examples of the patent in suit for which an amount of iron supported on the surface (in  $\mu\text{g}/\text{m}^2$ ) is indicated, none of the samples exemplified

provides such a value in the absence of an indication of the specific surface area of the particles. Furthermore, it is worth noting that the same calculation is valid regardless of the operational mode used for the synthesis of the particles, i.e. hydrolysis or neutralisation of a titanium compound in an aqueous solution containing the iron compound dissolved therein as for sample O or impregnation of different types of titanium dioxide particles with an iron compound as for samples A to D, F, G, I, K and N, despite the fact that different operational modes are not expected to provide the same degree of homogeneity of the iron compound within the titanium dioxide particles.

10. Consequently, the skilled person would inevitably come to the conclusion that the feature "said iron compound supported in an amount of 0.05 to 5,000  $\mu\text{g}$  in terms of Fe per square meter of the surface of said titanium oxide particles" does not define a concentration of iron on the surface of the titanium oxide particles, as construed by the Opposition Division, but is meant to define the ratio of the amount of iron contained in the titanium oxide particles, i.e. on the surface and in the volume of the particles, to the specific surface area of said particles. This does not mean, as reasoned by the Opposition Division, that all iron present in the particles must be present on the surface thereof, but merely that the particles are further defined by the ratio of iron (regardless of its distribution in the particles) to the specific surface area of the particles. Furthermore, it is not disputed that both values and their ratio can be easily determined by the skilled person, the specific surface area being

suitably measured as known in the art using the BET method, as is confirmed by D1 and D2 also in relation to the same type of compounds. Whether the specific surface area value of any titanium oxide particle to be determined might depend on the experimental settings and the conditions employed for the measurement which are not specified in the claims as granted is a clarity issue in relation to the limits of present claim 1. This ambiguity, if any, would not result from amendments made in opposition proceedings but would have been already present in the claims as granted. Furthermore, it would not prevent the person skilled in the art from carrying out the present invention, as the skilled person would be able to obtain an amount of iron per specific surface area of the titanium oxide particles ranging from 0.05 to 5 000 µg of Fe per square metre on the one hand by adjusting the amount of iron and on the other hand, based on his general knowledge in the present field, by varying the specific surface area of the titanium oxide particles, as determined by the selected method and experimental conditions, adjusting the conditions selected for hydrolysis or neutralisation, drying and, if required, firing. Finally, it is also contested that the titanium oxide particles comprising an iron compound which are taught in the patent in suit would exhibit photocatalytic activity to a degree suitable for synthesis of organic substances or decomposition of noxious materials under irradiation of ultraviolet rays.

11. Consequently, it follows from the above that the invention as defined in claim 1 according to the Main Request is sufficiently disclosed to be carried out by a person skilled in the art. The same holds true for

dependent claims 2 to 5, which merely define the preferred mineral acid, iron compound, amount of iron "per square meter of the surface area of the titanium oxide particles" and type of titanium oxide.

*Remittal*

12. The patent was revoked by the Opposition Division on the sole ground that present claim 1 lacked sufficiency of disclosure. Numerous other issues have yet to be examined and decided on. The Board accordingly deems it appropriate, exercising its discretion under Article 111(1) EPC, to remit the case to the department of first instance for further prosecution.



**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 for further prosecution on the basis of the claims according to the Main Request submitted during the oral proceedings before the Opposition Division.

The Registrar

The Chairman

S. Fabiani

J. Riolo