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Datasheet for the decision of 24 June 2010

Case Number: T 1764/06 - 3.3.07

Application Number: 01308838.0

Publication Number: 1199103

IPC: B01J 35/00

Language of the proceedings: EN

Title of invention:
Photocatalyst, process for producing the same and photocatalyst coating composition comprising the same

Applicants:
Sumitomo Chemical Company, Limited

Headword:
-

Relevant legal provisions (EPC 1973):
EPC Art. 54(1)

Keyword:
"Novelty (no) - Definition of a parameter feature representing the only difference from the prior art - Presumption of lack of novelty not displaced by evidence - Benefit of doubt not accorded (Main request)"

Decisions cited:
-

Catchword:
In a situation where the applicants have used an unusual parameter feature to define their product, which unusual parameter feature represents the only distinction over otherwise identical known products, and the applicants have decided not to provide evidence that the parameter feature as such represents a difference of the claimed products from the known products, no benefit of doubt can be accorded (Paragraph 2.12 of the Reasons)

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Case Number: T 1764/06 - 3.3.07

D E C I S I O N
of the Technical Board of Appeal 3.3.07
of 24 June 2010

Appellants: Sumitomo Chemical Company, Limited
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Decision under appeal: **Decision of the Examining Division of the European Patent Office posted 6 June 2006 refusing European application No. 01308838.0 pursuant to Article 97(1) EPC.**

Composition of the Board:

Chairman: S. Perryman
Members: G. Santavicca
D. Semino

Summary of Facts and Submissions

I. The appeal lies from a decision of the Examining Division refusing European patent application 01 308 838.0 (Publication Number EP 1 199 103 A2), concerning a "photocatalyst, process for producing the same and photocatalyst coating composition comprising the same". That decision was based on a set of Claims 1 to 12 faxed on 23 March 2006, in preparation for the oral proceedings held on 24 April 2006.

II. According to the decision under appeal, the catalysts illustrated in any of D1 (L. Palmisano et al, "*Surface properties of iron-titania photocatalysts employed for 4-nitrophenol photodegradation in aqueous TiO_2 dispersion*", Catalysis Letters 24 (1994), pages 303-315, J.C. Baltzer AG, Science Publishers) and D2 (J. Soria et al., "*Dinitrogen Photoreduction to Ammonia over Titanium Dioxide Powders Doped with Ferric Ions*", Journal of Physical Chemistry, vol. 95, 1991, pages 274-282, American Chemical Society) were prepared in a manner similar to that for preparing the claimed catalysts and were identical to the claimed catalysts in all respects other than Index X. As the applicants had not shown that the catalysts of any of D1 and D2 did not have an Index X of 0.2 or more, the subject-matter of Claim 1 of the then Main Request lacked novelty over D1 or D2. It was also pointed out in the decision under appeal that, should it be shown by appropriate comparative tests that the known catalysts, which had been prepared in a similar manner as the claimed catalysts, did not exhibit an Index X as claimed, it would be questionable whether the

application had sufficiently disclosed all the features for preparing the claimed products (Article 83 EPC).

III. In their statement setting out the grounds of appeal, the appellants enclosed copy of a further document (D7, Kazuhito Hashimoto et al, "*A History of the Study of Titanium Oxide Photocatalysts*", in "Photocatalyst: Fundamental, Material Development, and Application", pages 24-30, first impression of the first edition published on 27 May 2005, NTS, Tokyo, Japan) and submitted a new Main Request.

Claims 1 to 11 of the Main Request read as follows (compared to the claims as filed, additions of features are shown in bold, deletions in strike-through):

"1. A photocatalyst comprising a titanium oxide and a metal-containing compound other than titanium oxide on the surface of the titanium oxide, wherein the metal-containing compound is a metal oxide having an acid site ~~and~~**,** the photocatalyst has a BET specific surface area of ~~about~~ 55 m^2/g or larger, ~~or wherein the metal-containing compound is a basic metal-containing compound and the photocatalyst has an anatase-crystalline structure and an anatase-crystalline size of about 10nm or larger~~ **and the photocatalyst has an index X calculated by equation (I):**

X=B/A (I)

of 0.2 or more, wherein A represents an integrated value of absorbance within a wavelength range of from 220 nm to 800 nm along an ultraviolet-visible diffuse reflection spectrum of the photocatalyst, and B represents an integrated value of absorbance within a

wavelength range of from 400 nm to 800 nm along the ultraviolet-visible diffuse reflection spectrum."

"2. A photocatalyst according to claim 1, which contains the metal-containing compound in an amount of ~~about~~ 0.05% by mol**e or more and 10% by mole or less** in terms of the metal element of the compound, based on titanium in the photocatalyst."

"3~~5~~. A photocatalyst according to claim ~~4~~ 1 to 2, which has a BET specific surface area of ~~about~~ 300 m^2/g or smaller."

"4~~10~~. A process for producing a photocatalyst as claimed in any one of claims 1 to ~~9~~3, which comprises the steps of allowing a titanium oxide to come in contact with a solution or slurry of a metal-containing compound other than titanium oxide and/or a solution or slurry of a compound which makes the metal-containing compound by heating; and heating the resulting titanium oxide, to obtain a photocatalyst."

"5~~11~~. A process for producing a photocatalyst as claimed in any one of claims 1 to 3, which comprises the steps of putting in a sealed-container, a titanium oxide and at least one compound selected from a metal-containing compound and a compound which makes the metal-containing compound by heating; heating and evaporating the compound; and allowing the compound to adhere to the surface of the titanium oxide, to obtain a photocatalyst."

"6~~12~~. A process for producing a photocatalyst as claimed in any one of claims 1 to 3~~9~~, which comprises

the steps of placing a metal-containing compound on a titanium oxide, and treating the resulting titanium oxide having the metal-containing compound thereon with steam or carbon dioxide gas, to obtain a photocatalyst."

"7~~13~~. A process according to any one of claims 4~~10~~ to 6~~12~~, wherein the titanium oxide has an anatase-crystalline structure."

"8~~14~~. A process according to any one of claims 4~~10~~ to 7~~13~~, wherein the titanium oxide is obtained by a process comprising the steps of reacting a titanium compound with a base to obtain a reaction product at a temperature of 60°C or lower; and calcining the reaction product to obtain a titanium oxide."

"9~~15~~. A process according to any one of claims 4~~10~~ to 7~~13~~, wherein the titanium oxide is obtained by a process comprising the steps of calcining titanium oxysulfate in the presence of oxygen."

"10~~16~~. A photocatalyst coating composition comprising a photocatalyst as claimed in any one of claims 1 to 3~~9~~ and a solvent."

"11~~17~~. A material coated with a photocatalyst according to any one of claims 1 to 3~~9~~ or a photocatalyst coating composition according to claim 10~~16~~.".

IV. The Board, in a communication in preparation for oral proceedings, addressed the issues to be decided, in particular, in view of the feature "Index X ... of 0.2 or more ", the clarity of that definition, the relevant

sufficiency of the disclosure and the novelty of the photocatalyst so defined. Having regard to the decision under appeal, attention was drawn to the outstanding objections of lack of novelty over the photocatalyst disclosed by D1. Still concerning Index X, the Board indicated that the question arose whether the arguments of the applicants in connection with coloured and white titanium oxide were plausible in the absence of any evidence over the prior art, e.g. D1, nor was it understood why the anatase photocatalyst samples of D1 having the required BET surface area, acid sites, containing up to 5% of iron, including possible islands or coatings of Fe_2O_3, were still considered to be as white as untreated titanium oxide.

V. In response to the communication of the Board in preparation for oral proceedings, the appellants, by their letter dated 16 June 2010, announced that they no longer wished to participate to the oral proceedings and withdrew their request for oral proceedings. No comments whatsoever in reply to the outstanding issues raised in the communication of the Board were given.

VI. Oral proceedings were held on 24 June 2010 in the announced absence of the applicants appellants.

VII. In their statement setting out the grounds of appeal the appellants, *inter alia*, had argued that:

(a) The new Main Request only differed from the request underlying the decision under appeal by the deletion of Claim 4 and the consequential renumbering of the remaining claims. Hence, the new claim request was admissible.

(b) The conclusion of the Examining Division on lack of novelty was erroneous for three reasons: D1 and D2 were not clear and unambiguous disclosures of the catalysts as claimed; the evidence before the Examining Division showed that the prior UV photo-catalysts did not meet the requirement for Index X; in such a complex situation, without direct disclosure, the benefit of doubt should be given to the applicants.

(c) As regards the significance of Index X, it was important to consider that it was generally known that titanium oxide was white. Thus, the person skilled in the art of photocatalysis would note the colour of the material they were working with, if any. If none of D1 and D2 contained any reference to colour, this implied that the relevant materials were not coloured. In that respect, attention was drawn to D3 (G.C. Bond et al, "*Structure and Reactivity of Titania-supported Molybdenum and Tungsten Oxides*", Catalysis Today, 1(1987), 229-243, Elsevier Science Publishers B.V., Amsterdam), which mentioned explicitly the colour of a particular catalyst.

(d) As D1 and D2 did not mention that the treated titanium oxide had any colour, which implied that it was white, neither D1 nor D2 directly and unambiguously disclosed the requirement for Index X, which, if the material was white, would be less than 0.2.

(e) Nor had the catalyst samples of D1 and D2 been prepared according to a method as disclosed in the present application. For obtaining an Index X as defined, i.e. a coloration of the oxide material, it was necessary to incorporate a sufficient amount of the additional metal-containing compound by a process as described in the paragraph bridging pages 4 and 5 of the application as filed. This process involved the use of a stoichiometric excess of a base, which was responsible for the coloration and the ability of the resulting product to act as a visible light activated photocatalyst.

(f) Finally, the known catalysts were activated by ultraviolet (UV) rather than by visible light. D7 confirmed that D1, D2 and even D6 (O.A. Ileperuma, "*Photocatalysis Behaviour of Metal doped Titanium Dioxide, Studies on the Photochemical Synthesis of Ammonia on Mg/TiO_2 Catalyst Systems*", Applied Catalysis, 62(1990), pages L1-L5) did not concern visible-light activated photocatalysts.

(g) Therefore, the claimed subject-matter was novel.

(h) As regards sufficiency, the examples evidenced that it was possible to make the claimed catalysts and the skilled person had no difficulty in adapting the said examples to reproduce the claimed catalyst over the whole scope claimed.

VIII. The appellants had requested in writing that the impugned decision be set aside and a patent be granted on the basis of Claims 1 to 11 of the Main Request

submitted with the statement setting out the grounds of appeal (letter dated 9 October 2006). Furthermore, if the Board had any doubts as to inventive step or sufficiency of the disclosure, they requested the remittal of the case to the Examining Division, to safeguard their right to appeal.

Reasons for the Decision

1. The appeal is admissible.

Main Request

2. *Novelty*

2.1 The sole ground for refusal was the lack of novelty of the subject-matter as claimed in the request underlying the decision under appeal, having regard to D1 and D2.

The subject-matter of the present Main Request differs from that of the request underlying the decision under appeal only in the deletion of the then dependent Claim 4. Hence, the independent claims, in particular Claim 1, remain the same as those underlying the decision under appeal.

2.2 D1 concerns iron doped polycrystalline titania powders having photo-activity in catalytic processes (Introduction, page 303).

2.2.1 In particular, D1 discloses the preparation of iron doped samples by impregnation of pure TiO_2 (mainly anatase) with an aqueous solution containing Fe(III)

ions, their comparison with co-precipitated specimens, their characterization as well as the testing of their photoactivity in the photodegradation of 4-nitrophenol in aqueous dispersion (Introduction, page 304).

2.2.2 As regards the preparation of the impregnated samples, respectively having nominal concentration of 0.01, 0.02, 0.04, 0.1, 0.5, 1.0, 3.0 and 5.0 mol of iron per 100 mol of titanium, firstly a TiO_2 (mainly anatase) solid material was prepared by reacting an aqueous solution of $TiCl_3$ with an aqueous solution of ammonia added dropwise at room temperature with vigorous stirring and by filtering, washing, drying and calcining the obtained home prepared titania. That material was divided in different fractions and each of them was impregnated with an aqueous solution of the required amount of Fe^{3+} ions, then dried and calcined (page 304, Experimental, first paragraph).

2.2.3 The specimens were *inter alia* characterized for their phases, surface area, porosity as well as for their adsorption of the species ammonia, pyridine and methanoic acid (probe molecules) monitored by Fourier-Transform InfraRed (FTIR) spectroscopy technique, to assess the surface acidity and basicity (page 304, Introduction, second full paragraph; page 305, points 2.2 to 2.4).

2.2.4 As regards the characterization by X-Ray diffraction of the phases, samples with iron content lower than 3% (atomic) showed the lines of the home prepared titania, in particular intense peaks of anatase and some minor peaks of rutile, and no peak due to any other phase, whereas for iron loading higher than 3% peaks due to

hematite (Fe_2O_3) were also recorded (page 306, results and discussion, point 3.1). Also, it was found that the excess of iron produced layers or islands of inactive amorphous and/or crystalline material (Fe_2O_3 for instance) on the catalyst surface, which probably was not effective for the O_2 reduction and the oxidation of 4-nitrophenol (page 314, first full paragraph, last sentence; third full paragraph, last sentence).

2.2.5 As to the surface area (table 1 on page 306), the impregnated samples having nominal concentration of 0.1, 0.5, 1.0, 3.0 and 5.0 mol of iron per 100 mol of titanium developed each specific surface areas of 55 m^2/g or higher.

2.2.6 The characterization by FTIR spectroscopy of the adsorption of pyridine and ammonia (page 308, point 3.3) showed, in particular for the samples having nominal concentration of 0.5 and 5% mol of iron, the presence of surface Lewis acid sites (page 309, full paragraph, second sentence) and of surface Brønsted acid sites (page 310, point 3.3.2, second paragraph, fourth sentence).

2.2.7 Finally, the photo-reactivity-test (page 312, point 3.4) showed that the photo-activity became less and less significant as the iron content increased, i.e. that the presence of iron ions did not beneficially influence the photoactivity of pure TiO_2 (anatase) for the photodegradation of 4-nitrophenol, in the given experimental conditions. However, this had to be contrasted to e.g. the known essentiality of the presence of metal ion in gas-solid regime for the photoreduction of dinitrogen to ammonia (page 313,

first complete sentence; paragraph bridging pages 313 and 314). In other words, the samples were still photoactive but their activity depended on the reaction.

2.2.8 It follows from the above that D1 discloses a photocatalyst prepared according to a method as defined in claims 4 and 8 of the Main Request (*supra*) and comprising all the features of Claims 1 to 3 of the Main Request (*supra*) but the Index X.

2.3 According to Claim 1 (*supra*), the photocatalyst has an index X of 0.2 or more, calculated by equation X = B/A (I), wherein A represents an integrated value of absorbance within a wavelength range of from 220 nm to 800 nm along an ultraviolet-visible diffuse reflection spectrum of the photocatalyst, and B represents an integrated value of absorbance within a wavelength range of from 400 nm to 800 nm along the ultraviolet-visible diffuse reflection spectrum. Hence, the claimed photocatalyst also has a performance related characteristic expressed by a parameter feature.

2.4 In their letter dated 23 March 2006 (page 2, last two paragraphs), the applicants offered the following interpretation of Index X: "A" represented the amount of absorption of a broad wavelength range of light, including both the ultraviolet and the visible region; "B" represented the amount of absorption in the visible region of light; "Index X", which was equal to "B/A", therefore showed the proportion of ultraviolet-visible light that was absorbed in the visible region; the larger the Index X, the larger the proportion of light that was absorbed within the visible region; Index X thus related to the strength of the colour of the

titanium oxide; when the titanium oxide was white Index X was about 0.1; the claimed photocatalyst had an Index X of more than 0.2, i.e. it absorbed enough light within the visible region and was not white.

2.5 The Board notes that:

(a) None of the documents cited in the proceedings as representing the prior art ever mentions such an Index X. Not even D5 (Masakazu Anpo, "*Photocatalysis on titanium oxide catalysts, Approaches in achieving highly efficient reactions and realizing the use of visible light*", Catalysis Surveys *from Japan*, 1(1997), pages 169-179), which concerns the use of visible light in the photocatalysis on titanium oxide catalysts, ever mentions Index X.

(b) No evidence whatsoever that "Index X" was common general knowledge in the field of photo-catalysis or had already been disclosed or used has ever been offered either.

(c) D7, alleged to represent the history of the development of visible light photo-catalysis, does not mention either any Index X.

(d) The characterization of the photocatalysts disclosed by D1 does not include the assessment of the colour. Nor does D2 or D6.

2.6 Hence, the only expressed distinction between the definition of the claimed photocatalyst and the photocatalyst of D1 relies on a parameter feature that is not usual, let alone common, for the skilled person.

2.7 Furthermore, if the significance of Index X given by the applicants were retained, i.e. that it is related

to the strength of the colour of the titanium oxide, nothing of this would be found in the application as filed.

Instead, from D1 (page 314, first full paragraph, last sentence; fourth paragraph, last sentence), it is learnt that an excess of iron, as in samples having more than 3% mol of iron, *a fortiori* the sample having 5% mol, produces, on the catalyst, surface layers and/or islands of inactive amorphous and/or crystalline material such as hematite.

Hematite is generally known to be coloured, which fact must have an impact on the colour of the titanium oxide catalyst and on its capacity of absorbing visible light.

2.8 Also, the method of preparation of the samples illustrated in D1 is as defined in Claims 4 and 8 of the Main Request, and as described in the paragraph bridging pages 4 and 5 of the application as filed.

In particular, in D1, ammonia is added dropwise, hence until completion of the reaction.

As regards the loading, the examples of the present application illustrate 3% mol of metal. Concerning Index X, the present application shows that also the comparative samples obtained by impregnation of commercially available titanium oxide (which have not been shown to have been produced by the alleged excess of ammonia) attain an Index of as high as 0.2 (Comparative Example 2), with a loading of 3% mol.

Hence, no proven distinction appears to result from the method of preparation either, let alone from the alleged excess of ammonia, which further is not a feature required by any of the claims.

2.9 Summing up, the applicants have not provided any items of evidence showing that the samples of D1, e.g. those having a high iron content, do not attain an Index X as required. However, the onus of the proof in that respect lay on them, and no proven distinction has ever been shown.

2.10 Therefore, in these circumstances, the presumption that the claimed catalyst is not novel (as detailed throughout the examination and appeal proceedings), having regard to e.g. D1, has not been displaced by evidence.

2.11 Consequently, the condition required by Article 52 EPC (... provided that they are new ...) is not fulfilled by the catalysts defined in Claim 1, and a European patent cannot be granted on the Main Request.

2.12 As regards the invoked "benefit of doubt", to be accorded to the applicants in complex situations without direct disclosure, the position of the Board is as follows: in a situation where the applicants have used an unusual parameter feature to define their product, which unusual parameter feature represents the only distinction over otherwise identical known products, and the applicants have decided not to provide evidence that the parameter feature as such represents a difference of the claimed products from the known products, no benefit of doubt can be accorded.

Article 52 EPC requires that the invention (here, the claimed photocatalyst) be novel.

2.13 The above objections extend to the subject-matter defined in Claims 2 and 3 as well as to the method of preparation as defined in Claims 4 and 8.

2.14 Therefore, the Main Request is not acceptable.

3. *Conclusions*

The claimed subject-matter as defined in the Main Request does not fulfil the requirements of the EPC, so that no European patent can be granted.

Given the above decision, the Board need not review the decision under appeal on the question whether also the disclosure of D2 is novelty destroying.

Since the appeal fails on novelty, the Board need not give further details on the issues of fair basis for the claims (Article 123(2) EPC), clarity (Article 84 EPC) and sufficiency of the disclosure (Article 83 EPC) as mentioned in the communication of the Board in preparation for oral proceedings.

Order

For these reasons it is decided that:

The appeal is dismissed.

The Registrar: The Chairman:

S. Fabiani S. Perryman