BESCHWERDEKAMMERN DES EUROPÄISCHEN PATENTAMTS

BOARDS OF APPEAL OF THE EUROPEAN PATENT OFFICE

CHAMBRES DE RECOURS DE L'OFFICE EUROPEEN DES BREVETS

Internal distribution code:
(A) [ ] Publication in OJ
(B) [ ] To Chairmen and Members
(C) [ ] To Chairmen
(D) [X] No distribution

Datasheet for the decision of 20 September 2011

Case Number: T 2254/10 - 3.4.03

Application Number: 99942386.6

Publication Number: 1048084

IPC: H01L 31/06, B32B 9/04, H01L 25/04, H01L 51/20, H01L 51/30, H01L 31/0224, H01L 27/146, H01L 27/142

Language of the proceedings: EN

Title of invention:
Organic photosensitive optoelectronic device

Applicant:
THE TRUSTEES OF PRINCETON UNIVERSITY

Opponent:
-

Headword:
-

Relevant legal provisions:
-

Relevant legal provisions (EPC 1973):
EPC Art. 56

Keyword:
"Inventive step (no)"

Decisions cited:
-

Catchword:
-

Europäisches Patentamt | European Patent Office | Office européen des brevets

Beschwerdekammern | Boards of Appeal | Chambres de recours

Case Number: T 2254/10 - 3.4.03

D E C I S I O N
of the Technical Board of Appeal 3.4.03
of 20 September 2011

Appellant: (Applicant)
THE TRUSTEES OF PRINCETON UNIVERSITY
P.O. Box 36
Princeton
NJ 08544-0636 (US)

Representative:
Kiriczi, Sven Bernhard
Schneiders & Behrendt
Mühlthaler Strasse 91d
D-81475 München (DE)

Decision under appeal:
Decision of the Examining Division of the European Patent Office posted 20 August 2010 refusing European patent application No. 99942386.6 pursuant to Article 97(2) EPC.

Composition of the Board:

Chairman: G. Eliasson
Members: R. Q. Bekkering
P. Mühlens

Summary of Facts and Submissions

I. This is an appeal against the refusal of application 99 942 386 for lack of clarity, Article 84 EPC 1973, (main request) and lack of an inventive step, Article 56 EPC 1973, (first and third auxiliary requests) over document

D7: Bonnet D., Luke U.: "Organic Solar Cells - An Experimental Study", 13th European Photovoltaic Solar Energy Conference, vol. 2, 23-27 October 1995, Nice, France, pages 1685-1688.

II. At oral proceedings before the board, the appellant applicant requested that the decision under appeal be set aside and a patent granted on the basis of the following:

Main request:

Claims 1 to 16 of the main request filed on 30 June 2010 with the letter dated 25 June 2010;

First auxiliary request:

Claim 1 of auxiliary request 1 filed with the letter dated 19 August 2011;
Claims 2 to 16 as for the main request;

Second auxiliary request:

Claims 1 to 15 of auxiliary request 2 filed on 30 June 2010 with the letter dated 25 June 2010.

Furthermore, the appellant requested that, in case the board intended to remit the case again, the case be remitted to a different examining division.

III. Claim 1 of the main request reads as follows:

"*A series stacked organic photosensitive optoelectronic device comprising:*
a substrate having a proximal surface and a distal surface; and
a plurality of organic photosensitive optoelectronic subcells in superposed relation with each other and with said proximal surface of said substrate, each of said subcells having a transparent cathode, a transparent anode and at least a photoconductive layer, each of said cathode and anode being an electrode layer or a charge transfer layer, each of said subcells sharing at least one electrode layer or charge transfer layer with an adjacent subcell,
wherein said plurality of organic photosensitive optoelectronic subcells are electrically connected in series."

IV. Claim 1 of the first auxiliary request corresponds to claim 1 of the main request with the following addition:

"*wherein a charge transfer layer delivers charge carriers from one subsection of an optoelectronic device to the adjacent subsection*".

V. Claim 1 of the second auxiliary request corresponds to claim 1 of the main request with the following addition:

"*wherein thicknesses of photoconductive organic layers vary between subcells or subcells have photoconductive organic materials with different absorption characteristics*".

VI. Reference is made to the following further documents:

D1: Zhang J. et al., "*Photovoltaic properties of porphyrin solid films with electric-field induction*", Thin Solid Films, vol. 284-28, 15 September 1996, pages 596-599

D8: Peumann P. et al., "*Small Molecular Weight Organic Thin-Film Photodetectors and Solar Cells*", Journal of Applied Physics, vol. 93(7), 1 April 2003, pages 1-31

D9: Hiramoto M. et al., "*Effect of Thin Gold Interstitial-layer on the Photovoltaic Properties of Tandem Organic Solar Cell*", Chemistry Letters 1990, The Chemical Society of Japan, pages 327-330

D10: EP 1 336 995 A

D12: Coutts T.J. and Meakin J.D (Eds.), "*Current Topics in Photovoltaics*", Academic Press, London, 1985, pages 36 and 37

D13: Rand B. et al., "*Long-range absorption enhancement in organic tandem thin-film solar cells containing*

silver nanoclusters", Journal of Applied Physics, vol. 96(12), 15 December 2004, pages 7519-7526

D14: Karl N. et al., "*Efficient Organic Photovoltaic Cells. The Role of Excitonic Light Collection, Exciton Diffusion to Interfaces, Internal Fields for Charge Separation, and High Charge Carrier Mobilities*", Molecular Crystals and Liquid Crystals, vol. 252, 1994, pages 243-258

VII. The appellant in substance provided the following arguments:

Document D7 did not disclose a charge transfer layer. The double Ag and Cu film was not a proper layer but rather a collection of islands and provided charge recombination rather than charge transfer, as could be inferred from documents D8, D9 and D13. Furthermore, document D7, or any of the other cited prior art, failed to disclose a transparent (in particular ITO) top electrode. Moreover, it was clear from D7 that tandem cells were a dead end in view of their poor performance. Accordingly, the skilled person would not even consider any further modifications. Hence, the subject-matter of claim 1 according to the main request, as well as that of the auxiliary requests which contained further limitations, was both new and inventive over the cited prior art.

Reasons for the Decision

1. The appeal is admissible.

2. *Main request*

2.1 *Novelty*

2.1.1 *Document D7*

Document D7 discloses organic solar cells having a heterojunction diode using Cu-phthalo-cyanine (Cu-Pc) as p-conductor and di-methyl-perylene (MPP) as n-conductor (cf pages 1685, 1686, Sections 1, "*Introduction*" and 2, "*Experimental procedures*" and figure 1). An ITO-coated glass is used as substrate onto which the p-n film stack is formed. In particular, D7 discloses, as further optimisation, a tandem cell comprising stacked cells with a transparent contact between both diodes, converting a hole current into an electron current (cf page 1687, Section 3.2, "*Cell optimisation*", subsection B, "*Tandem cells*" and figure 6). A double film of 20 Å Ag and 20 Å Cu is used, silver being supposed to make an ohmic contact to the n-MPP and Cu being supposed to make an ohmic contact to p-CuPc. The result in this case, according to D7, is that "*indeed the voltages add up to 0.9 V*", the stacked cells being connected in series.

Although the appellant argued that figure 6 of D7 showed that the tandem cell only produced an open circuit voltage of 0.45 V, in the board's opinion there can be no doubt to the skilled reader of D7 that the value "*0.5*" indicating the scale of the horizontal axis

is incorrect and should read "*1.0*". In this case, the open circuit voltage of the tandem cell structure as shown in figure 6 is 0.9 V, which corresponds to what is stated in the text and is consistent with the voltages disclosed for the elementary cells.

Accordingly, document D7 discloses, in the terminology of claim 1,
- a series stacked organic photosensitive optoelectronic device comprising:
- a substrate having a proximal surface and a distal surface; and
- a plurality of organic photosensitive optoelectronic subcells in superposed relation with each other and with said proximal surface of said substrate,
- each of said subcells having a cathode, an anode and at least a photoconductive layer,
- each of said cathode and anode being an electrode layer or a charge transfer layer,
- each of said subcells sharing at least one electrode layer or charge transfer layer with an adjacent subcell,
- wherein said plurality of organic photosensitive optoelectronic subcells are electrically connected in series.

2.1.2 The appellant argued that the double film of 20 Å Ag and 20 Å Cu of D7 was not a charge transfer layer as required in claim 1. In particular, it was argued that, as could be seen from eg documents D9, D8 and D13, the double film of D7 in fact was not a layer but merely a collection of islands or clusters. Moreover, charge recombination rather than charge transfer took place at the double film of D7.

2.1.3 *Documents D9, D8 and D13 referred to by the appellant*

2.1.3.1 *Document D9*

Document D9 discloses a tandem organic solar cell, each unit cell comprising a layer of metal-free phthalocyanine (H_2Pc) and a layer of perylene tetracarboxylic derivative (Me-PTC) forming a pn-junction (cf page 327; figure 2). The tandem cell comprises an ultra-thin Au interstitial-layer (< 3nm) between the unit cells, providing an ohmic contact between the H_2Pc of the front cell and the Me-PTC of the back cell, resulting in a doubling of the open circuit voltage V_{oc} (page 328, first to third paragraph; figure 5). According to D9, "*Since the pigment film is polycrystalline and its surface is not uniform (Fig. 6), deposited thin Au layer on the pigment film seems to have an island structure*" (page 329, last paragraph).

Moreover, as to the tandem cell's functioning, and in particular to that of the Au layer, D9 indicates that "*the effective recombination between electrons in Me-PTC of the back cell and holes in H_2Pc of the front cell has to take place to flow the photocurrent through the tandem cell*" (page 329, last paragraph). According to D9, "*At only the Au sites, the photogenerated holes in the front cell and the photogenerated electrons in the back cell can recombine. Outside of the Au sites, electrons and holes were separated each other due to the p-n junction formed between H_2Pc and Me-PTC*" (page 329, last paragraph to page 330, first paragraph).

2.1.3.2*Document D8*

Document D8, published after the filing date of the present application and cited by the appellant, discloses an organic tandem thin-film solar cell consisting of two stacked cells connected in series separated by an ultrathin silver layer. The silver layer, deposited with an average thickness of 5 Å, is found to form isolated clusters (page 22, left-hand column, second paragraph to right-hand column, first paragraph).

Concerning the functioning of the tandem cell, D8 discloses that "*deposition of two cells in series leads to formation of an inversely oriented heterojunction between the acceptor layer of one cell and the donor layer of the adjacent cell. To prevent carrier pile-up at the inverse heterojunction, a contact layer is inserted between the individual cells, providing a recombination site for electrons and holes approaching from adjacent cells. The contact layer provides a site for charge recombination, resulting in the alignment of the Fermi levels from adjacent cells and ensuring a minimal loss of photovoltage*" (page 18, right-hand column, second paragraph) and "[*instead,*] *an ultrathin layer of metal nanoclusters is sufficient to provide sites for the unpaired photogenerated charges to recombine with near unity efficiency under 1 sun illumination (~100mW/cm^2)*" (page 22, right-hand column, first paragraph).

2.1.3.3*Document D13*

Document D13, also published after the filing date of the present application and cited by the appellant, discloses an organic tandem thin-film solar cell consisting of two stacked cells connected in series separated by a silver nanocluster layer (pages 7519, right-hand column, last paragraph, to page 7520, left-hand column, first paragraph; figure 1). The silver nanocluster layer is deposited to a thickness of 1 nm (page 7521, right-hand column, first paragraph; page 7525, table I). Moreover, according to D13, "*the charge recombination layer in a tandem organic PV cell consists of a thermally evaporated, random array of Ag clusters of various sizes, shapes and spacings*" (page 7523, right-hand column, second paragraph).

As to the tandem cell operation and in particular to that of the Ag nanocluster layer, D13 discloses that "*upon light absorption, excitons are formed in both photovoltaic subcells. After dissociation at a DA interface, the hole in PV 1 and electron in PV 2 are collected at the adjacent electrodes. To prevent buildup of charge within the cells, the electron in PV 1 and hole in PV 2 diffuse to the metal nanocluster layer where they recombine. The attraction of the initial charge to the nanoparticle is primarily a result of image charge effects. Once the metal particle is singly charged, Coulomb attraction of the free counter charge leads to rapid recombination at the Ag surface*" (page 7519, right-hand column, last paragraph to page 7520, left-hand column, first paragraph; figure 1).

2.1.4 As to the appellant's argument above, it is noted that none of the above documents D9, D8 and D13 disclose a double film of Ag and Cu as provided in D7. Moreover, even if the double film of Ag and Cu of D7 were to be formed of islands or clusters, all of the above documents make nonetheless reference to a "*layer*", be it a Au layer having an island structure (D9), an ultrathin Ag layer (D8) or an Ag nanocluster layer (D13).

Last but not least, the application description itself as originally filed, when referring to document D9, refers to the interstitial Au layer as a "*charge transfer layer*" (original description, page 13, lines 16 to 26).

The fact that this part of the description has been deleted in subsequently filed amendments can also not impart a different meaning to the expression "*charge transfer layer*" as used in the application, as this would render the amendment inadmissible under Article 123(2) EPC. Moreover, the deletion can also not be considered a correction under Rule 88 EPC 1973, as it is not immediately evident that a mistake occurred.

Accordingly, the double film of document D7 constitutes a layer as required in claim 1 under consideration.

2.1.5 Moreover, it is clear from straightforward physical considerations that in order to prevent charge from building up at respective sides of the double film of D7, inhibiting proper operation of the tandem cell, the photogenerated charge carriers in the form of electrons and holes, diffusing from respective sides to the

double film, must recombine. Evidently, this requires charge carriers to be transferred across the thickness of the double film to recombine with opposite charge carriers. Accordingly, the double film of D7 constitutes a charge transfer layer as required by claim 1 under consideration.

The explanations provided in document D9, D8 and D13 discussed above, insofar as the double Ag and Cu film of D7 can be compared with the Au or Ag layers provided in these documents, confirm that the photogenerated charge carriers diffusing from either side to the double film recombine at the metal surface. Even if the layer consists of islands or nanoclusters of metal, this still requires charge carriers to be transferred across the thickness of these islands or nanoclusters to recombine with opposite charge carriers.

Accordingly, also following these considerations, the double film of D7 constitutes a charge transfer layer as per claim 1 under consideration.

2.1.6 At the oral proceedings the appellant provided further explanations based on a number of slides as to the underlying physical mechanism distinguishing a charge transfer layer as claimed from the above prior art. It was submitted that in accordance with the application, both holes and electrons would be transferred across the charge transfer layer, which was presented as a relative thick layer, eg a 100-400 nm thick ITO layer as described in the applicant's specification (cf slides submitted at the oral proceedings). It is noted, however, that the fact that holes were alleged to transfer through the thick ITO layer casts doubts onto

the soundness of these explanations. Regarding the above prior art, it was argued that in the case of relative thin island, charge carrier recombination based on image charge effects, rather than charge transfer, took place. It is however noted that even following the offered model based on image charges (cf slides submitted at the oral proceedings), the positive charge of a hole present on one side of the island ultimately is transferred across the island to the electron on the opposite side of the island.

Accordingly, even by these explanations offered by the appellant, a layer of islands or nanoclusters as provided in the above prior art would constitute a charge transfer layer.

Whether the layer is actually called a charge transfer layer or a charge recombination layer would, thus, appear to be merely a matter of semantics rather than of any technical substance, the underlying physical mechanism involving both charge transfer and charge recombination being identical.

This is corroborated by the fact that, as already noted above, the application itself when referring to document D9, refers to the interstitial Au layer as a "*charge transfer layer*" (original description, page 13, lines 16 to 26).

2.1.7 Moreover, the double film of Ag and Cu of document D7, providing the contact between the two elementary cells, is disclosed to be transparent (page 1687, Section B, "*Tandem cells*").

2.1.8 Finally, it is considered implicit from D7 that for the tandem cell the same ITO-coated glass is used as substrate, as is described for the single cell (cf page 1685, right-hand column, last paragraph; figure 1).

2.1.9 Not disclosed in D7, on the other hand, is a transparent top electrode, the only top electrode described being that of the single cell, which consists of an array of Ag or Al dots (cf page 1685, right-hand column, last paragraph; figure 1).

2.1.10 Accordingly, the subject-matter of claim 1 differs from D7 in that the top electrode of the series stacked device is transparent.

The subject-matter of claim 1, thus, is new over document D7.

2.1.11 In document D9, the top electrode is a 20 nm Au layer and, thus, not transparent.

Accordingly, the subject-matter of claim 1 is also new over document D9.

2.2 *Inventive step*

2.2.1 Taking document D7 as the closest prior art, the subject-matter of claim 1, thus, differs in that the top electrode of the device is transparent.

The effect hereof, for example, is that light incident from the top can also contribute to the photovoltaic process.

Accordingly, the objective problem-to-be-solved relative to document D7 is allowing light incident from the top to contribute to the photovoltaic process.

This problem-to-be-solved *per se* is well known to a person skilled in the art working in the technical field at issue of photovoltaic devices.

The solution as claimed is the provision of a transparent top electrode.

This solution is considered to be part of the common general knowledge of the person skilled in the art.

2.2.2 The appellant argued that in view of technical difficulties encountered in applying a transparent electrode layer consisting of Indium tin oxide (ITO) on organic photosensitive material, the skilled person would be lead away from the claimed solution.

However, it is noted that the solution as claimed merely involves a transparent top electrode in general and is not limited to any specific material such as ITO.

Moreover, it is noted that at any rate no technical prejudice is apparent that would have prevented the person skilled in the art from at least trying the use of a widely employed material such as ITO, generally a first choice material for electrodes in the field at issue for its rather exceptional combination of transparency and high conductivity.

The appellant specifically referred to document D14 acknowledged in the application as originally filed, as

evidence that the skilled person would be dissuaded from using ITO. According to the application, however, this document merely indicates that the ITO electrode was found to be very rarely photoactive (cf page 9, second paragraph). Photoactivity is, however, not required in tandem cells such as provided in D7 where ITO is used as a top electrode providing an ohmic contact.

Moreover, at any rate a number of other documents (see document D1, figure 1 and corresponding description; document D10, figure 1 and corresponding description; see also application, page 12, second paragraph) confirm that the provision of ITO on organic photosensitive material is possible, so that the appellant's allegation of a technical prejudice against such an arrangement must be dismissed.

2.2.3 The appellant argued in this respect that documents D1 and D10 could not render the provision of an ITO top electrode in the tandem cell of D7 obvious, as the cell of D1 was not delivering any sizeable power and D10 concerned a photoemissive device.

It is, however, noted that, as stated above, in the board's judgement ITO is so common in the technical field at issue, that it would be obvious to the skilled person to try to use it for the top electrode, or any other electrode or contact layer requiring transparency and high conductivity for that matter.

Documents D1 and D10, even if concerned with related photosensitive device and not specifically with tandem cells, do not demonstrate the existence of the

technical prejudice against the use of ITO on organic photosensitive material invoked by the appellant.

It should be noted in this respect that the mere fact that technical difficulties are encountered in the prior art when applying a certain measure does not as such imply the presence of a technical prejudice. A technical prejudice can only be said to exist when the skilled person would not even consider carrying out experiments to determine whether the measure works. This is clearly not the case for ITO as a transparent electrode material.

Finally, it is noted that if indeed the provision of ITO on organic photosensitive material had presented difficulties in the prior art, not the mere provision of an ITO electrode but if anything a full solution as to how to proceed to overcome these difficulties could be seen as providing an inventive contribution. Claim 1 under consideration is silent on any of this.

2.2.4 Moreover, the appellant argued that the skilled person, based on the poor performance of the tandem cell of D7, would consider tandem cells a dead end and not even consider further developments.

The appellant relies in particular on figure 6 of document D7, showing an open circuit voltage of the tandem cell of only 0.45 V. However, as already stated above, in the board's opinion it is clear to the skilled reader of D7 as a whole that the open circuit voltage of the tandem cell is 0.9 V. The performance deterioration of the tandem cell as calculated by the

appellant is thus not correct (see slides presented at the oral proceedings).

Moreover, there can be no question of tandem cells being perceived in the art as a "dead end" or indeed any technical prejudice against such cells.

According to D7, for the tandem cell "*the current is too low due to insufficient matching of both diodes and the reduced transparency of the intermediate contact. Nevertheless this procedure shows a way to increased performance*" (page 1687, right-hand column, first paragraph). The dismissal of this statement by the appellant as undue speculations is not justified. In fact, contrary to what is argued by the appellant, in D7 the tandem cell is perceived as promising and even ways for improvement, in the form of proper matching of both diodes and improved transparency of the contact, are suggested.

It is, moreover, noted that the fact that the low electrical current produced by tandem cells is generally known to be caused by the cells being connected in series. Tandem cells, on the other hand provide the known advantage of higher output voltages, desirable for certain applications, as well as higher conversion efficiency. The skilled person, thus, rather than being confronted with any technical prejudice as argued by the appellant, merely is presented with the known advantages and disadvantages of tandem cells with respect to single cells.

Nothing else is in fact the case in the application itself, where it is indicated that "*although the high*

series resistance of photoconductive organic materials inhibits use of subcells in a series configuration for high power applications, there are certain applications, for example, in operating liquid crystal displays (LCD), for which a higher voltage may be required, but only at low current and, thus, at low power levels" (page 21, lines 5 to 8).

Neither could the appellant's argument convince that it was indicative of tandem cells being considered a dead end, or indeed the inventive merit of the application, that a long time lapsed between the publication of prior art relating to tandem cells and the present application, in which no activity in this field took place. Document D7 was published only about three years before the priority date of the application.

2.2.5 Accordingly, the subject-matter of claim 1 of the main request is obvious to a person skilled in the art and, thus, lacks an inventive step in the sense of Article 56 EPC 1973.

The appellant's main request is, therefore, not allowable.

3. *First auxiliary request*

Claim 1 of the first auxiliary request corresponds to claim 1 of the main request with the following addition:

"*wherein a charge transfer layer delivers charge carriers from one subsection of an optoelectronic device to the adjacent subsection*".

In the discussion above with respect to claim 1 of the main request the charge transfer layer is understood to deliver charge carriers from one subsection of an optoelectronic device to the adjacent subsection.

Accordingly, the subject-matter of claim 1 of the first auxiliary request is obvious to a person skilled in the art for the same reasons given above for claim 1 of the main request. The subject-matter of claim 1 of the first auxiliary request, thus, lacks an inventive step in the sense of Article 56 EPC 1973.

The appellant's first auxiliary request is, therefore, not allowable either.

4. *Second auxiliary request*

Claim 1 of the second auxiliary request corresponds to claim 1 of the main request with the following addition:

"*wherein thicknesses of photoconductive organic layers vary between subcells or subcells have photoconductive organic materials with different absorption characteristics*".

Both measures above provide for an optimisation of the tandem cell in terms of current matching and/or absorption characteristics. The objective problem-to-be solved relative to D7 may be formulated accordingly as providing such an optimisation.

Both solutions as claimed to the above problem-to-be solved of varying the thicknesses of the photoconductive layers between subcells as well as of

providing photoconductive materials with different absorption characteristics are generally known (cf document D12, pages 36-37) and also specifically known for organic tandem cells (see eg D9, page 327, last paragraph and page 329, first paragraph).

Accordingly, the subject-matter of claim 1 of the second auxiliary request is also obvious to a person skilled in the art and, thus, lacks an inventive step in the sense of Article 56 EPC 1973.

The appellant's second auxiliary request is, therefore, also not allowable.

5. The appellant's request for remitting the case to a different examining division, accordingly, need not be considered.

Order

For these reasons it is decided that:

The appeal is dismissed.

Registrar: Chair:

S. Sánchez Chiquero G. Eliasson