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Datasheet for the decision of 8 March 2018

Case Number: T 1046/16 - 3.3.09

Application Number: 02750605.4

Publication Number: 1374320

IPC: H01L51/00, H01L51/20

Language of the proceedings: EN

Title of invention:
MATERIALS AND DEVICES FOR BLUE PHOSPHORESCENCE BASED ORGANIC LIGHT EMITTING DIODES

Applicants:
THE TRUSTEES OF PRINCETON UNIVERSITY
UNIVERSITY OF SOUTHERN CALIFORNIA

Headword:

Relevant legal provisions:
EPC Art. 84, 123(2), 56

Keyword:
Claims - clarity (yes)
Amendments - allowable (yes)
Inventive step - (yes)

Decisions cited:

Catchword:

Beschwerdekammern

Boards of Appeal

Chambres de recours

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Case Number: T 1046/16 - 3.3.09

D E C I S I O N
of Technical Board of Appeal 3.3.09
of 8 March 2018

Appellant: (Applicant 1)
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Decision under appeal:
Decision of the Examining Division of the European Patent Office posted on 14 December 2015 refusing European patent application No. 02750605.4 pursuant to Article 97(2) EPC.

Composition of the Board:

Chairman W. Sieber
Members: M. O. Müller
F. Blumer

Summary of Facts and Submissions

I. European patent application No. 02 750 605.4, filed on 13 March 2002 as international application PCT/US02/07492 in the name of the Trustees of Princeton University and the University of Southern California was refused by decision of the examining division.

II. The application as filed contains two independent claims 1 and 13, which read as follows:

"1. An emissive layer of an organic light emitting device, said emissive layer comprising:
a wide gap host material;
a charge carrying dopant material, present as a dopant in the wide gap host material; and
a phosphorescent dopant material, present as a dopant in the wide gap host material;
wherein the charge carrying dopant material comprises a hole transporting material and the phosphorescent dopant material comprises an electron transporting material."

"13. An emissive layer of an organic light emitting device, said emissive layer comprising:
a wide gap host material;
a charge carrying dopant material, present as a dopant in the wide gap host material, and
a phosphorescent dopant material, present as a dopant in the wide gap host material;
wherein the charge carrying dopant material comprises an electron transporting material and the phosphorescent dopant material comprises a hole transporting material."

III. The examining division cited the following document in its communications and decision:

D1: DE 44 28 450 A1.

IV. The examining division's decision was based on a main request and first and second auxiliary requests. Essentially, claims 1 and 2 of these requests differed from claims 1 and 13 of the application as filed (point II above) in that the term "wide gap host material" was amended to "host material" (main request and first auxiliary request) and in that a new definition for the host material was inserted (all requests).

The examining division did not admit the main request under Rule 137(3) EPC, since the deletion of the expression "wide gap" in relation to the host material did not meet the requirements of Article 123(2) EPC.

It found the first auxiliary request not to be allowable, since the deletion of the expression "wide gap" violated Article 123(2) EPC and the new definition of the host material did not comply with Article 84 EPC.

The second auxiliary request was held not to be allowable, since the definition of the host material was considered not to meet the requirements of Article 84 EPC.

In an obiter dictum, the examining division considered the subject-matter of all requests not to be inventive in view of the teaching of D1.

V. This decision was appealed by the joint applicants (hereinafter the appellants). With the statement of grounds of appeal, the appellants filed a main request and first to third auxiliary requests as well as

E1: Declaration of Prof. Thompson, signed 21 April 2016, including his curriculum vitae and a list of publications, patents and invited talks;

E2: C. W. Tang et al., Applied Physics Letters, volume 51, 1987, pages 913 to 915;

E3: C. W. Tang et al., Journal of Applied Physics, volume 65, 1989, pages 3610 to 3616; and

E4: "Electroluminescence I - Semiconductors and Semimetals", G. Mueller (ed.), volume 64, 2000, 33 pages.

VI. On 10 November 2017, the board communicated its preliminary opinion to the appellants.

VII. With their letter dated 8 February 2018, the appellants filed new requests 1 to 4 as well as new page 22 and withdrew the then pending requests under the condition that the new requests were considered in lieu of the then pending requests and not merely rejected as being late filed.

VIII. With their letter dated 28 February 2018, the appellants filed request 5 and

E5: Declaration of Prof. Thompson signed on 27 February 2018.

IX. On 8 March 2018, oral proceedings were held before the board, during which the appellants filed a main request, claims 1 and 2 of which read as follows:

"1. An emissive layer of an organic light emitting device, said emissive layer comprising
a wide gap host material,
a charge carrying dopant material, present as a dopant in the wide gap host material, and
a phosphorescent dopant material, present as a dopant in the wide gap host material,
wherein the charge carrying dopant material is a hole transporting material and the phosphorescent dopant material is an electron transporting material,
wherein said phosphorescent dopant material emits from a triplet excited state of an organic molecule in said phosphorescent dopant material, and
wherein the lowest triplet state energy level of the wide gap host material (T_W) is higher than the lowest triplet state energy level of the electron transporting material (T_{ET}) ($T_W > T_{ET}$), and the lowest triplet state energy level of the hole transporting material (T_{HT}) is higher than the lowest triplet state energy level of the electron transporting material (T_{ET}) ($T_{HT} > T_{ET}$),
wherein the HOMO-LUMO energy gap of the wide gap host material is greater than the HOMO-LUMO energy gap of both the hole and electron transporting materials,
the HOMO level of the wide gap host material is lower than the HOMO level of any of the dopant materials as well as any adjacent layers that are in direct physical contact with the wide gap host material, and

the LUMO level of the wide gap host material is higher than the LUMO level of any of the dopant materials, as well as any adjacent layers that are in direct physical contact with the host material."

"2. An emissive layer of an organic light emitting device, said emissive layer comprising
a wide gap host material,
a charge carrying dopant material, present as a dopant in the wide gap host material, and
a phosphorescent dopant material, present as a dopant in the wide gap host material,
wherein the charge carrying dopant material is an electron transporting material and the phosphorescent dopant material is a hole transporting material,
wherein said phosphorescent dopant material emits from a triplet excited state of an organic molecule in said phosphorescent dopant material, and
wherein the lowest triplet state energy level of the wide gap host material (T_W) is higher than the lowest triplet state energy level of the hole transporting material (T_{HT}) ($T_W > T_{HT}$), and the lowest triplet state energy level of the electron transporting material (T_{ET}) is higher than the lowest triplet state energy level of the hole transporting material (T_{HT}) ($T_{ET} > T_{HT}$),
wherein the HOMO-LUMO energy gap of the wide gap host material is greater than the HOMO-LUMO energy gap of both the hole and electron transporting materials,
the HOMO level of the wide gap host material is lower than the HOMO level of any of the dopant materials as well as any adjacent layers that are in direct physical contact with the wide gap host material, and

the LUMO level of the wide gap host material is higher than the LUMO level of any of the dopant materials, as well as any adjacent layers that are in direct physical contact with the host material."

X. The appellants' position in the written and oral proceedings, in so far as relevant to the present decision, was as follows:

In view of the definition of the HOMO and LUMO levels of the host material in claims 1 and 2, the feature that this material was a wide gap host material was clear. According to the two claims, the HOMO and LUMO levels of the host material had to fulfill three conditions, which were those graphically depicted in figure 4 of the application as filed. By meeting these three conditions, the host material was inevitably such that it had a wide gap that rendered the material inert in the sense that it did not carry any charges when the OLED was operated.

The HOMO and LUMO levels of the adjacent layer(s) mentioned in claims 1 and 2 did not raise any clarity issue either. It was clear that they referred to the highest HOMO and the lowest LUMO of the material(s) contained in this (these) layers.

The definition of the HOMO and LUMO levels of the host material was based on page 16, line 19, to page 17, line 16, of the application as filed. The remaining features of claims 1 and 2 were also disclosed in the application as filed. These claims thus met the requirements of Article 123(2) EPC.

The subject-matter of claims 1 and 2 was inventive in view of D1. It differed from this document in that according to these claims emission came from the triplet excited state of an organic molecule, rather than from a metal atom and in that the phosphorescent dopant was at the same time an emitter and an electron or hole transporting material. The problem solved in view of D1 was the provision of an OLED with a simplified structure. D1 did not suggest any phosphorescent dopant that at the same time functioned as an emitter and charge carrier. It in fact taught away from it, since it required a phosphorescent dopant that did not influence the transport properties of the charge carrying materials.

XI. The appellants requested that the decision under appeal be set aside and a patent be granted on the basis of claims 1 to 21 of the main request filed during the oral proceedings before the board on 8 March 2018 or on the basis of the claims of any of requests 1 to 4 filed with their letter dated 8 February 2018 or the claims of request 5 filed with their letter dated 28 February 2018.

Reasons for the Decision

Main request

1. The invention

1.1 Claims 1 and 2 refer to an emissive layer of an organic light emitting device (OLED) comprising an electron transporting material, a hole transporting material and a wide gap host material.

1.2 As can be deduced from page 9, lines 10 to 23, in conjunction with page 1, lines 3 to 7, and page 11, lines 19 to 24, of the application as filed, the "wide gap" of the host material renders it inert, in the sense that it does not carry any charges when the OLED is operated. This characteristic is the crucial feature of the invention described in the application as filed. By way of this characteristic, excitons are preferentially formed at the dopant site, rather than in the host, and thereby lead to the blue phosphorescence of the OLED aimed at in the application as filed.

2. Article 84 EPC

2.1 The term "wide" is a relative term that does not precisely define the size of the gap. Hence, the feature of the host material being a wide gap host material as such is unclear.

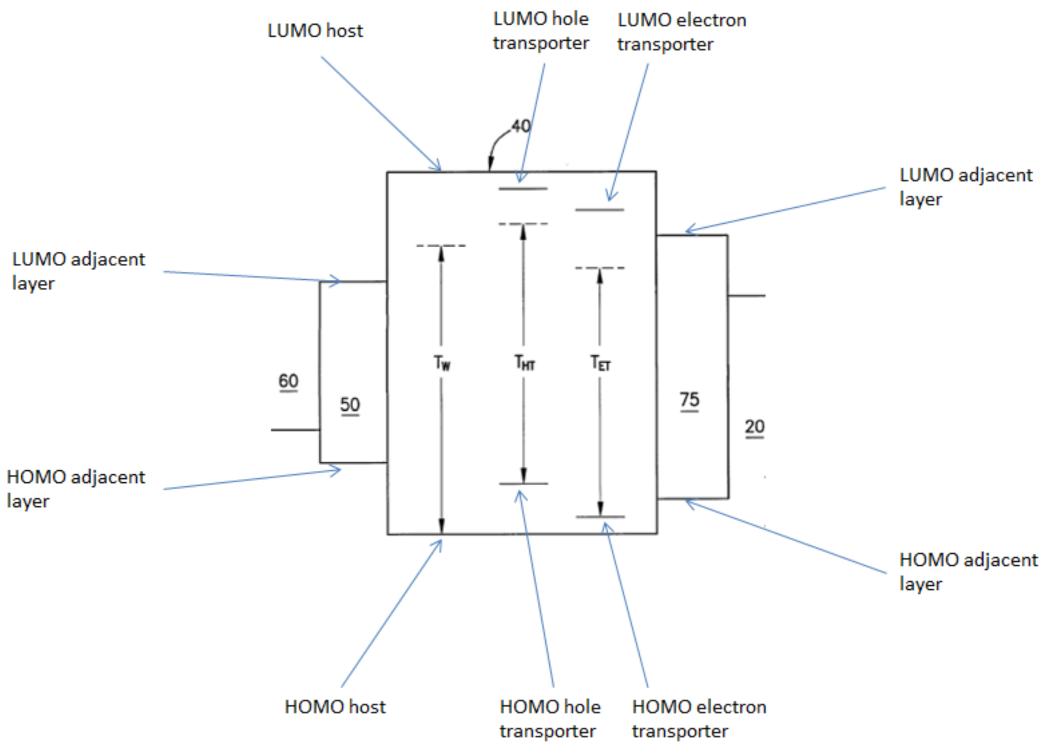
2.1.1 However, in the main request now on file, claims 1 and 2 do not define the host material only by the feature of having a wide gap but additionally contain three conditions the host material must fulfill:

Firstly, the HOMO-LUMO energy gap of the wide gap host material must be greater than the HOMO-LUMO energy gap of both the hole and electron transporting materials.

Secondly, the HOMO level of the wide gap host material must be lower than the HOMO level of any of the electron and hole transporting materials as well as any adjacent layers that are in direct physical contact with the wide gap host material.

Thirdly, the LUMO level of the wide gap host material must be higher than the LUMO level of any of the electron and hole transporting materials, as well as any adjacent layers that are in direct physical contact with the host material.

2.1.2 These three conditions of claims 1 and 2 are graphically shown in figure 4 of the application as filed. This figure is reproduced below together with explanations of the meaning of its individual parts:



As can be seen in this figure, the energy gap between the HOMO and LUMO of the host (the two horizontal lines of the perimeter of box 40) is greater than the corresponding gaps of the electron and hole transporting materials (horizontal solid lines in the middle and right-hand part of box 40). Hence, the first condition of claims 1 and 2 is fulfilled.

The HOMO level of the host (lower horizontal line of the circumference of box 40) is lower than the HOMO levels of the electron and hole transporting materials and that of any adjacent layer (lower horizontal lines of the perimeters of boxes 50 and 75), i.e. the second condition of claims 1 and 2 is fulfilled.

Lastly, the LUMO of the host (upper horizontal line of the perimeter of box 40) is higher than the LUMO level of the electron and hole transporting materials and that of any adjacent layer (upper lines of the perimeters of boxes 50 and 75), i.e. the third condition of claims 1 and 2 is also fulfilled.

2.1.3 As the host material has the widest HOMO-LUMO gap of all materials, it is a wide gap material. As explained by Prof. Thompson during the oral proceedings, and as stated in his declaration E5 (point 5), the inevitable consequence is that, when the OLED is operated, the host material does not carry any charges, so it is inert. More specifically, if the HOMO level of the host material is the lowest of all HOMO levels, no hole will be created in this level, since no electron will be extracted from it and transferred to any HOMO level of the electron or hole transporting material or adjacent layer. Furthermore, if the LUMO of the host material is the highest of all LUMO levels, no negative charge will be created in it, since no electron will be transferred to it from any LUMO level of the electron or hole transporting material or any adjacent layer. Lastly, if the gap between the HOMO and LUMO levels of the host material is the largest of all gaps, no charge transfer will take place within this material, i.e. between its HOMO and LUMO level.

The board is aware that according to page 17, lines 12 to 16, of the application as filed, the fulfillment of the three conditions of claims 1 and 2 leads only "typically", and thus arguably not always, to a wide gap or, in other terms, inert host material. As however confirmed by Prof. Thompson, the term "typically" in the quoted passage is erroneous, since, as set out above, meeting the three conditions inevitably, rather than typically, results in a wide gap or inert host material.

2.1.4 The feature of the host material being a wide gap or inert material is thus defined in a non-ambiguous way by the three conditions present in claims 1 and 2. This feature is thus not unclear, so no deficiency under Article 84 EPC arises.

2.2 In its preliminary opinion, the board had raised the question what was meant by the HOMO and LUMO levels of any adjacent layer in claims 1 and 2. As observed by the board during the oral proceedings, this appeared to be unclear in cases where this layer contained more than one material and thus more than one HOMO and LUMO level.

As however explained by Prof. Thompson during the oral proceedings, the levels of the adjacent layer(s) referred to in claims 1 and 2 can only be the highest HOMO and the lowest LUMO level of the adjacent layer(s), since, when the OLED is operated, a hole or electron (if any) is injected into these two levels. The host will only be inert if the hole or electron injected into the adjacent layer cannot be transferred to it. This means that the HOMO of the host must be lower than the highest HOMO and the LUMO of the host must be higher than the lowest LUMO of the adjacent layer(s). The skilled person reading claim 1 would thus know that the HOMO and LUMO of any adjacent layer in claims 1 and 2 refers to its highest HOMO and lowest LUMO. There is thus no ambiguity.

2.3 The board is also convinced that the remaining features of claims 1 and 2 are clear. The same applies for all remaining claims. The claims of the main request thus meet the requirements of Article 84 EPC.

3. Article 123(2) EPC

3.1 Claim 1

3.1.1 The feature

> "An emissive layer of an organic light emitting device, said emissive layer comprising a wide gap host material, a charge carrying dopant material, present as a dopant in the wide gap host material, and a phosphorescent dopant material, present as a dopant in the wide gap host material, wherein the charge carrying dopant material is a hole transporting material and the phosphorescent dopant material is an electron transporting material"

is based on claim 1 as filed.

The amendment of "comprises" to "is" (underlined portions above) is based on page 5, lines 1 to 18, page 16, lines 10 to 11 and 14 to 15, and page 18, lines 21 to 24, of the application as filed.

3.1.2 The feature in claim 1 "wherein said phosphorescent dopant material emits from a triplet excited state of an organic molecule in said phosphorescent dopant material" is based on page 8, lines 18 to 19 of the application as filed.

3.1.3 The feature in claim 1 "wherein the lowest triplet state energy level of the wide gap host material (T_W) is higher than the lowest triplet state energy level of the electron transporting material (T_{ET}) ($T_W > T_{ET}$), and the lowest triplet state energy level of the hole transporting material (T_{HT}) is higher than the lowest triplet state energy level of the electron transporting material (T_{ET}) ($T_{HT} > T_{ET}$)" is based on claim 2 as filed.

3.1.4 The feature in claim 1 that

> "the HOMO-LUMO energy gap of the wide gap host material is greater than the HOMO-LUMO energy gap of both the hole and electron transporting materials,
> the HOMO level of the wide gap host material is lower than the HOMO level of any of the dopant materials as well as any adjacent layers that are in direct physical contact with the wide gap host material, and
> the LUMO level of the wide gap host material is higher than the LUMO level of any of the dopant materials, as well as any adjacent layers that are in direct physical contact with the host material"

differs from the disclosure on page 16, line 21 to page 17, line 5, of the application as filed only in that the term "inert host material" in this passage has been replaced by the term "wide gap host material". As set out in point 2.1.3 above, both terms mean the same thing in the application as filed, namely that the host material does not receive a charge when the OLED is operated. Hence, the above feature of claim 1 is based on the application as filed.

3.2 Claim 2

3.2.1 The feature

> "An emissive layer of an organic light emitting device, said emissive layer comprising
> a wide gap host material, a charge carrying dopant material, present as a dopant in the wide gap host material, and a phosphorescent dopant material, present as a dopant in the wide gap host material, wherein the charge carrying dopant material is an electron transporting material and the phosphorescent dopant material is a hole transporting material"

is based on claim 13 as filed.

The amendment of "comprises" to "is" (underlined portions above) is based on page 5, lines 1 to 18, page 16, lines 10 to 11 and 14 to 15, and page 18, lines 21 to 24, of the application as filed.

3.2.2 The feature in claim 2 "wherein said phosphorescent dopant material emits from a triplet excited state of an organic molecule in said phosphorescent dopant material" is based on page 8, lines 18 to 19, of the application as filed.

3.2.3 The feature in claim 2 "wherein the lowest triplet state energy level of the wide gap host material (T_W) is higher than the lowest triplet state energy level of the hole transporting material (T_{HT}) ($T_W > T_{HT}$), and the lowest triplet state energy level of the electron transporting material (T_{HT}) is higher than the lowest triplet state energy level of the hole transporting material (T_{ET}) ($T_{ET} > T_{HT}$)" is based on claim 14 as filed.

3.2.4 The remaining features of claim 2 are identical to those of claim 1 and thus are equally based on the application as filed.

3.3 Claim 3 is based on claim 10 as filed.

3.4 Claims 4 and 5 are based on claims 22 and 25 as filed, except that "oxidiazole" has been changed to "oxidazole". This however simply corrects an obvious error, as is evident from the formula in claim 5, which contains an oxidazole rather than an oxidiazole ring.

3.5 Claims 6 to 21 are based on claims 9 and 21, 11 and 23, 12 and 24, 3 and 15, 4 and 16, 6 and 18, 7 and 19, 8 and 20, 26 and 46, 38 and 59, 39 and 60, 40 and 61, 41 and 62, 42 and 63, 43 and 64 and 45 and 66 of the application as filed.

4. Inventive step in view of D1

4.1 In an obiter dictum, the examining division considered the first auxiliary request then on file not to be inventive in view of D1.

4.2 The present application is directed to phosphorescent OLEDs (page 1, lines 3 to 7).

4.3 D1 is directed to organic electroluminescent devices (column 1, lines 3 to 4). Emission in these devices results from triplet excitons (column 2, lines 53 to 60), which implies phosphorescence. Therefore, in the same way as the application as filed, D1 refers to phosphorescent OLEDs. D1 can thus be considered to be a possible starting point for assessing inventive step.

4.4 D1 discloses an OLED comprising a complex of a rare earth metal with organic ligands as phosphorescent dopant, one or more organic electron transporting materials and, optionally, one or more organic hole transporting materials (column 2, lines 23 to 51, and claim 1 of D1).

The optional organic hole transporting material can be present in a matrix of polymethyl methacrylate or bisphenol A-polycarbonate (column 5, lines 47 to 52), corresponding to the host material of claims 1 and 2.

The emission in D1 comes from the metal atom of the rare earth metal complex (column 8, lines 52 to 61, of D1).

The concentration of the rare earth metal complex should not exceed 20 mol%, in order not to influence the transport properties of the charge carrying organic polymers, since these complexes mostly are insulators (column 6, lines 54 to 58).

The OLED of example 1 of D1 comprises a hole transporting layer made of poly(vinylcarbazole) and an electron transporting layer made of butyl-PBD doped with 10 wt% of the rare earth metal complex europium cinnamate. The OLED of example 2 of D1 comprises a hole transporting layer made of triphenyl amine, an electroluminescent layer made of terbium benzoate and an electron transporting layer made of PPD.

4.5 The subject-matter of claims 1 and 2 differs from D1 in that the emission comes from the triplet excited state of an organic molecule present in the phosphorescent dopant, rather than from a metal atom. A further difference is that according to claims 1 and 2, the phosphorescent dopant is an electron transporting material (claim 1) or a hole transporting material (claim 2). In D1, by contrast, the phosphorescent dopant and the electron and hole transporting materials are different materials.

4.6 During the oral proceedings, the appellants explained that the problem to be solved in view of D1 was the provision of an OLED with a simplified structure. This problem is derivable from page 9, lines 14 to 23, of the application as filed, where it is stated that the invention is directed to OLEDs having simple structures.

4.7 This problem is solved in view of D1. In this document, the emitting rare earth metal complex is separate from the hole and electron transporting material and in fact should not even influence their transport properties (column 6, lines 54 to 58). By contrast, in the emissive layer of claims 1 and 2, the phosphorescent dopant fulfils two functions, namely that of phosphorescent emission and electron or hole transport (claims 1 and 2 and page 9, lines 7 to 8). This means that one less layer is needed than in D1. The resulting OLED has a simpler structure than in D1.

4.8 D1 does not suggest any phosphorescent dopant that at the same time functions as an emitter and charge carrier. D1 in fact teaches away from such a dopant, since, as set out above, it requires a phosphorescent dopant that does not influence the transport properties of the charge carrying materials. Therefore, the subject-matter of claims 1 and 2, and by the same token of all remaining claims, is inventive in view of D1 itself.

5. Since the decision of the examining division is silent on the further documents cited in the international and European supplementary search report and the third party observation, the board finds it appropriate to remit the case to the examining division 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 examining division for further prosecution on the basis of claims 1 to 21 of the main request filed during the oral proceedings before the board on 8 March 2018.

The Registrar: The Chairman:

M. Cañueto Carbajo W. Sieber

Decision electronically authenticated