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Datasheet for the decision of 15 October 2012

Case Number: T 0203/10 - 3.3.03

Application Number: 02765394.8

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IPC: C08F 226/02, C08G 61/12, C09K 11/06, H05B 33/14

Language of the proceedings: EN

Title of invention:
Phosphor light-emitting compound, phosphor light-emitting composition, and organic light emitting element

Applicant:
NIPPON HOSO KYOKAI
Showa Denko K.K.

Headword:
-

Relevant legal provisions:
EPC Art. 54, 56, 84, 123(2)

Keyword:
"Clarity: no (main request); yes (auxiliary request 1)"
"Amendments - added subject-matter: no (auxiliary request 1)"
"Novelty: yes (auxiliary request 1)"
"Inventive step: yes (auxiliary request 1)"

Decisions cited:
-

Catchword:
-

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Case Number: T 0203/10 - 3.3.03

D E C I S I O N
of the Technical Board of Appeal 3.3.03
of 15 October 2012

Appellant I: (Applicant 1)
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Decision under appeal:
Decision of the Examining Division of the European Patent Office posted 7 September 2009 refusing European patent application No. 02765394.8 pursuant to Article 97(2) EPC.

Composition of the Board:

Chairman: B. ter Laan
Members: O. Dury
C.-P. Brandt

Summary of Facts and Submissions

I. The appeal by the applicants lies against the decision of the examining division posted 7 September 2009 to refuse European patent application N° 02 765 394.8.

II. The application as filed was based on 31 claims of which claims 1, 2, 11 und 13 read:

"1. A neutral organic polymeric phosphorescent compound emitting phosphorescence and used in an organic light-emitting device, characterized in that

a phosphorescent unit being a repeat unit for emitting phosphorescence and

a carrier transporting unit being a repeat unit for transporting a carrier

are comprised."

"2. The phosphorescent compound as claimed in claim 1, characterized in that a repeat number m for the phosphorescent unit(s) and a repeat number n for the carrier transporting unit(s) satisfy a relationship of $m < n$."

"11. The phosphorescent compound as claimed in any of claims 1 through 10, characterized in that a phosphorescent site of the phosphorescent unit is a monovalent group or a divalent group of a complex with a transition metal or a rare earth metal."

"13. The phosphorescent compound as claimed in any of claims 1 through 12, characterized in that a carrier transporting site of the carrier transporting unit comprises at least one kind of groups selected from

the group of groups consisting of monovalent groups of carbazole, monovalent groups of tertiary amine, monovalent groups of imidazole derivatives, monovalent groups of triazole derivatives, monovalent groups of oxadiazole derivatives, divalent groups of styrene, and divalent groups of fluorene and

the group of groups in which the groups are substituted with a substituent."

Claims 3-10, 12 and 14-16 were dependent claims directed to embodiments of claim 1. Claims 17-26 dealt with phosphorescent compositions comprising a phosphorescent compound according to claims 1-16. Claims 27-30 were directed to organic light-emitting devices comprising the phosphorescent compound or the phosphorescent composition according to claims 1-26. Claim 31 was directed to a display apparatus in which each pixel of the display screen consists of a light-emitting device of any of claims 27-30.

Furthermore, the passages on original page 32, lines 14-18 and 26-31 read, respectively, as follows:

"For example, by introducing three kinds of phosphorescent units radiating in blue, green, and red respectively to one compound in an appropriate proportion, a phosphorescent compound for white luminescence can be obtained."

"Furthermore, the blue color of the luminescent color mentioned herein is such that a peak wavelength in an emission spectrum is in 400 through 490 nm. Likewise, the green color is such that the peak wavelength is in

490 through 570 nm, and the red color is such that the peak wavelength is in 570 through 700 nm."

III. During the examination procedure the following documents were either cited in the contested decision (D1-D4), in a third party observation (A1-A3) or indicated as particularly relevant documents in the international search report (B1-B3):

D1: US 2001/0 015 432
D2: EP-A-0 992 564
D3: EP-A-1 006 169
D4: Molecularly doped polymer light emitting diodes utilizing phosphorescent Pt(II) and Ir(III) dopants, Lamansky et al., Organic Electronics 2, No. 1 (March 2001), pages 53-62
A1: EP-A-1 489 111
A2: US 2001/0 053 462
A3: EP-A-0 735 055
B1: JP 2002-293 830 A
B2: JP 2001-151 868 A
B3: EP-A-0 825 207.

IV. The decision under appeal was based on the sole request of the applicant comprising a set of claims filed with letter of 27 July 2009 together with amended pages 21, 25 and 56 of the description filed with letter of 28 March 2008, the remaining pages of the application as filed remaining unamended. The examining division refused the application holding, *inter alia*, that:

(a) the subject-matter claimed did not satisfy the requirements of Art. 84 EPC, because

(i) the definition of compounds or subunits by their colour was vague;

(ii) it was not clear "how to differentiate between different colors of different subunits in a molecule".

(iii) a reference to the description was not sufficient to render the claims as such clear.

(b) Regarding Art. 56 EPC, the subject-matter of claim 1 differed from the closest prior art D1 in that the polymer was neutral and colour indications were specified. The problem to be solved was identified as finding alternative phosphorescent Iridium complexes to those of D1. Considering that such compounds were generally known, in particular from D4, that their incorporation into the polymer of D1 was straightforward and that it was usual in the art to make organic light-emitting devices emitting in white by combining different red, green and blue emitting substances as shown e.g. in A2, the opposition division decided that the subject-matter of claim 1 was not inventive.

V. On 16 November 2009, the applicant (appellant) lodged an appeal against the above decision. The prescribed fee was paid on the same day. In its statement of grounds of appeal filed on 14 January 2010 the appellant requested that the decision of the opposition division be set aside and a patent be granted on the basis of the main request filed therewith. A test report was simultaneously filed.

VI. During the oral proceedings held on 15 October 2012, the appellant filed a new main request (12 claims) and two auxiliary requests (11 and 10 claims, respectively) replacing all former requests.

Claim 1 of the main request read as follows (additions are indicated in **bold** and deletions as ~~strikethrough~~, both as compared to claim 1 of the application as filed):

"1. A neutral organic polymeric phosphorescent compound emitting phosphorescence ~~and used~~ **for use** in an organic light-emitting device, ~~characterized in that~~ **comprising** ~~a~~ phosphorescent repeat unit**s** for emitting phosphorescence and a carrier transporting repeat unit for transporting a carrier ~~are comprised~~,

characterized in that the phosphorescent repeat units consist of three kinds radiating in blue with a spectral peak wavelength in 400 through 490 nm, green with a spectral peak wavelength in 490 through 570 nm, and red with a spectral peak wavelength in 570 through 700 nm,

wherein

a phosphorescent site of the phosphorescent repeat unit is a monovalent group or a divalent group of a complex with a transition metal or a rare earth metal and

a carrier transporting site of the carrier transporting repeat unit comprises at least one kind of group selected from

the group consisting of monovalent groups of carbazole, monovalent groups of tertiary amine, monovalent groups of imidazole derivatives, monovalent groups of triazole derivatives, monovalent groups of

oxadiazole derivatives, divalent groups of styrene, and divalent groups of fluorine and

groups in which these groups are substituted with a substituent."

Claim 1 of auxiliary request 1 read as follows:

"1. A ~~neutral~~ **non-ionic** organic polymeric phosphorescent compound emitting phosphorescence ~~and used~~ **for use** in an organic light-emitting device, ~~characterized in that~~ **comprising** ~~a~~ phosphorescent repeat unit**s** for emitting phosphorescence and a carrier transporting repeat unit for transporting a carrier ~~are comprised~~,

characterized in that a repeat number m for the phosphorescent unit(s) and a repeat number n for the carrier transporting unit(s) satisfy a relationship of m < n, and

characterized in that the phosphorescent repeat units consist of three kinds radiating in blue with a spectral peak wavelength in 400 through 490 nm, green with a spectral peak wavelength in 490 through 570 nm, and red with a spectral peak wavelength in 570 through 700 nm, and the phosphorescent compound radiates in white, wherein

the phosphorescent sites of the phosphorescent repeat units are a monovalent group or a divalent group of a complex with a transition metal or a rare earth metal and

a carrier transporting site of the carrier transporting repeat unit comprises at least one kind of group selected from

the group consisting of monovalent groups of carbazole, monovalent groups of tertiary amine,

monovalent groups of imidazole derivatives, monovalent groups of triazole derivatives, monovalent groups of oxadiazole derivatives, divalent groups of styrene, and divalent groups of fluorine and

groups in which these groups are substituted with a substituent."

Claims 2-4 of the first auxiliary request were directed to embodiments of claim 1, claims 5-7 to compositions comprising a compound according to claims 1-4, organic light-emitting devices comprising the phosphorescent compound or the phosphorescent composition according to claims 1-7. Claim 11 was directed to a display apparatus in which each pixel of the display screen **comprises** a light-emitting device of any of claims 8-10.

Auxiliary request 2 is not relevant for the present decision.

For each of those requests, amended pages 21, 25 and 56 filed with letter of 28 March 2008 were withdrawn, thereby returning to the version of the application as filed.

VII. The appellant's arguments that are relevant to the present decision may be summarised as follows:

Main request

(a) The subject-matter of claim 1 was derivable from the combination of claims 1, 11 and 13 together with the passages on page 32, lines 14-18 and 26-31 of the application as filed (Art. 123(2) EPC).

(b) No explanations were provided regarding the question of clarity (Art. 84 EPC) related to

- the term "neutral" of claim 1;
- the absence in independent claim 1 of the feature "m < n" as defined in claim 2;
- the expression "a phosphorescent site of the phosphorescent repeat unit is".

Auxiliary request 1

Art. 123(2) EPC

(c) The subject-matter of claim 1 was derivable from the combination of claims 1, 2, 11 and 13 together with the passages on page 32, lines 14-18 and 26-31 of the application as filed.

(d) The subject-matter of claims 2-11 was supported by the combination of each of claims 3, 4, 12, 21, 22, 23, 27 and 29-31 as originally filed, respectively, with the same passages of the application as filed identified above for claim 1, whereby the dependency of the claims was adapted accordingly.

Art. 84 EPC

(e) The skilled person knew that the radiation of the monomers used to build up the polymeric phosphorescent compound defined in claim 1 would not be modified upon polymerisation. Hence, the expression "the phosphorescent units (…) consists of three kinds radiating in blue (…), green (…) and red (…)" was to be read as meaning that the

phosphorescent monomers making up the polymeric compound radiated in blue, green and red as recited in claim 1.

Art. 54 EPC

(f) None of the documents cited in the proceedings disclosed a non-ionic, polymeric phosphorescent compound comprising three phosphorescent units radiating in blue, green and red, those units being a complex as defined in claim 1. Hence, the claimed subject-matter was novel.

Art. 56 EPC

(g) Starting from D1 as the closest prior art, the problem to be solved was to provide a phosphorescent compound emitting white luminescence and having improved quantum yield of luminescence as compared to the compounds of D1. Example 6-4 of the application together with the test report filed with the statement of grounds of appeal showed that that problem was indeed solved. Neither in D1, nor in any of the other cited documents was there a hint to solve the problem by modifying the teaching of D1 in such a way as to arrive at the subject-matter according to any of claims 1-11.

VIII. The appellant (applicant) requested that the decision under appeal be set aside and that a patent be granted on the basis of the main request or the auxiliary requests 1 or 2, all requests filed during the oral proceedings on 15 October 2012.

IX. The Board announced its decision at the end of the oral proceedings.

Reasons for the Decision

1. The appeal is admissible.

Main request

2. Clarity

2.1 Pursuant to Art. 84 EPC the claims, which define the subject-matter for which protection is sought, should be clear.

2.1.1 The term "neutral" (second word of claim 1) is used in the present application in order to characterise "non-ionic" compounds (see page 16, lines 28-29 of the application as filed).

However, as disclosed e.g. in D1 (second sentence of paragraph [0022]), ionic compounds comprising an equal number of negative and positive charges are also considered as being "neutral" in the present technical field. Besides, the term "neutral" as commonly used in chemistry has a further completely different meaning not related to the non-ionic character of a compound, namely in relation to the pH of a chemical compound.

The term "neutral" according to claim 1 has, thus, not a clear meaning.

2.1.2 Giving the claims, on the basis of the description (here e.g. page 16, lines 28-29), a specific meaning that is not reflected by the actual wording of the claims is, certainly at the examination stage, also not in accordance with the provisions of Art. 84 EPC.

2.1.3 The compounds defined in claim 1 contain three phosphorescent units radiating in blue, green and red. The expression "wherein **a** phosphorescent site of **the** phosphorescent repeat unit is a monovalent … group of a complex …" (emphasis by the Board), renders it unclear whether each of the red, blue and green phosphorescent repeat units has to be a complex as defined in claim 1 or if it is sufficient that at least one of those repeat units is such a complex.

2.1.4 For those reasons, the scope of claim 1 is not clearly defined and the skilled person is not in a position to determine whether or not he is working within or outside the claims.

2.2 The requirement of Art. 84 EPC, that the claims should be clear, is further read as meaning not only that a claim from a technical point of view must be comprehensible, but also that it must define clearly the object of the invention, i.e. indicate all the essential features thereof.

According to page 31, lines 11-21 of the application as filed, should the repeating number of the phosphorescent unit(s) be smaller than the repeating number of the carrier transporting unit(s) (i.e. should the criteria "m < n" according to claim 2 not be fulfilled), "emission of the luminescence is suppressed

by concentration quenching". This statement implies that the condition "m < n" is an essential technical feature of the invention. Considering that that feature is not present in claim 1, the requirements of Art. 84 EPC are also for that reason not met.

2.3 The main request is, thus, not allowable.

Auxiliary request 1

3. Amendments

3.1 Apart from some editorial amendments, claim 1 corresponds to the combination of claims 1, 2, 11 and 13 together with the passages on page 32, lines 14-18 and 26-31 of the application as filed with the following amendments:

(a) replacement of "neutral" by "non-ionic";

(b) replacement of "and use" by "for use";

(c) amendment of "**a** phosphorescent site of the phosphorescent repeat unit **is** …" by "**the** phosphorescent site**s** of the phosphorescent repeat unit**s are**" (emphasis by the Board).

3.1.2 Amendment (a) finds its basis in the passage on page 16, lines 28-29 of the application as filed: "… non-ionic, that is, neutral polymer.".

3.1.3 It is derivable from the whole application as filed that the intended use for the polymeric phosphorescent compounds claimed was the production of an organic light-emitting device. Amendment (b) is further supported by page 1, lines 5-11 and by page 3, lines 20-27 of the application as filed. Those statements of

general nature are applicable to any embodiment encompassed by the application as filed, including the embodiments according to present claims 1-11.

3.1.4 Amendment (c) imposes that each of the phosphorescent sites of the blue, green and red repeating units should be a complex of a transition metal or a rare earth metal as defined in claim 1. Considering that the application as filed, in particular the description and the examples, does not disclose any other kind of phosphorescent units than those now defined in claim 1, the subject-matter now being claimed does not extend beyond the content of the application as filed.

3.1.5 Claims 2-10 correspond to the combination of each of claims 3, 4, 12, 21, 22, 23, 27 and 29-30 as originally filed, respectively, with the same passages of the application as filed identified above for claim 1 (for claim 5, see in particular the combination of claims 21 and 17 as originally filed), whereby the dependency of the claims was adapted accordingly. The basis for claim 11 can be found in original claim 31 in combination with page 15, line 30, the word "includes" being considered equivalent to "comprises".

3.2 The requirements of Art. 123(2) EPC are therefore met.

4. Clarity

All the objections raised either by the Board or in the contested decision by the Examining Division have been taken into account by the amendments made. The Board is satisfied that the requirements of Art. 84 EPC are now met.

5. Novelty

5.1 D1 discloses a light emitting device comprising at least one emitting layer comprising an iridium complex (claim 6). The iridium complexes specified in claim 4 and paragraphs [0020]-[0021] are ionic compounds with an overall neutral charge. Compounds I-18, I-19 and I-20 (page 5) in particular illustrate three ionic organic polymeric phosphorescent compounds comprising a single phosphorescent repeat unit comprising an iridium complex and a carrier transporting unit either in the main chain (I-18, I-19) or in a side chain (I-20).

According to D1, paragraph [0018], the phosphorescent compound may carry more than one ligand. However, no polymer compound comprising the specific three different ligands radiating in red, green and blue as defined in present claim 1 is disclosed in D1.

5.2 D4 discloses doped polymeric organic light-emitting diodes (OLED) comprising phosphorescent complexes of platinum(II) or iridium(III) dispersed in a polymeric carrier (poly(N-vinylcarbazole)) (abstract; page 54, left column; full sections 3-4; Figs. 1 and 4). D4 does not disclose polymeric compounds comprising simultaneously carrier transporting units and phosphorescent unit(s), and thus, also not the combination of green, red and blue phosphorescent units according to claim 1.

5.3 A2 discloses a light-emitting device comprising an anode, an organic compound layer containing at least one light-emitting layer, and a cathode, wherein the at least one light-emitting layer contains two or more

different kinds of light-emitting materials, and at least one of the two or more light-emitting materials is an orthometallated complex (claim 1). The metal may be iridium (claim 5). The materials of A2 are however not polymers and are used as dopants in combination with other light emitting materials to provide white light organic light-emitting devices (claims 1-6 and 9-12; paragraphs [0018] and [0023]; examples 1 and 3). A2 therefore does not disclose *polymeric* materials comprising a complex of a metal, and thus also the combination of three phosphorescent units and a carrier transporting unit according to present claim 1.

5.4 B1 is a Japanese application that was published after the filing date of the present application, so that it is not valid prior art according to Art. 54(2)(3) EPC.

5.5 None of the other cited documents discloses a non-ionic, polymeric compound comprising three phosphorescent units radiating in blue, green and red, those units being a complex as defined in present claim 1.

5.6 The subject-matter of claim 1 as well as that of claims 2-11, which depend on it, is therefore novel.

6. Inventive step

6.1 Closest prior art

6.1.1 The present application relates to phosphorescent compounds, phosphorescent compositions and organic light-emitting devices as well as a display apparatus comprising said compounds. Such compounds are known from D1, which was considered as closest prior art by

both the appellant and the examining division. There is no reason to deviate from that view. In particular, any of compounds I-18, I-19 and I-20 represents a promising starting point.

6.1.2 A1 is a European patent application, which was published on 22 December 2004 in accordance with Art. 158(3) EPC. It corresponds to the international application published on 2 October 2003 as WO 2003/080687. Considering that both applications were published after the filing date of the present application (30 August 2002), none of them qualifies as prior art to be taken into account for inventive step (Art. 56 EPC, second sentence).

6.2 Problem to be solved

According to the application as filed (page 3, lines 11-27; page 4, lines 7-17; page 17, lines 1-3), the problem to be solved is to provide an organic polymeric phosphorescent material having improved emission efficiency.

During the oral proceedings before the Board the appellant further defined the problem to be solved, in reference to the closest prior art, as residing in providing an organic polymeric phosphorescent material having improved quantum yield of luminescence. That reformulation of the problem to be solved is derivable from the application as filed (see e.g. page 3, lines 11-27; page 36, line 24 to page 37, line 1) and is, thus, allowable.

6.3 Solution

The solution to the above-defined problem resides in the non-ionic organic polymeric phosphorescent compounds defined in claim 1. Those compounds differ from the polymeric phosphorescent compounds I-18 to I-20 of D1 in that:

- they are non-ionic;
- they comprise three repeating units radiating in blue, green and red as defined in claim 1.

6.4 Success of the solution - Reformulation of the problem

6.4.1 Example 6.4 of the application as filed discloses the preparation of a phosphorescent compound according to claim 1, which emits white light and is suitable for use in OLEDs.

6.4.2 The test report provided by the appellant with the statement of grounds of appeal shows that the quantum yield of a light emitting device using an ionic polymer compound is lower than that of the light-emitting device using a non-ionic compound according to claim 1.

6.4.3 However, page 31, lines 21-32 of the application as filed teaches that the emission efficiency depends on the respective amounts of phosphorescent units to the total number of phosphorescent and carrier units (ratio m/(m + n), wherein m and n are defined as in claim 1 and that the emission efficiency falls down when the amount of phosphorescent units is too small. As the ratio m/(m + n) is not present in claim 1 and hence claim 1 poses no restriction to that ratio, from the above-cited statement in the application as filed it

can be concluded that the problem identified above by the appellant is not credibly solved over the whole scope of the claims. The appellant did not provide any evidence or argument to refute that objection.

6.4.4 Therefore, the problem effectively solved has to be reformulated in a less ambitious manner, as being that of providing further, alternative organic polymeric phosphorescent material for organic light emitting devices.

6.5 Obviousness

6.5.1 It remains to be decided whether or not it was obvious to solve the above-identified problem by modifying the teaching of D1 in such a way as to arrive at the subject-matter of claim 1.

6.5.2 D1 neither provides a suggestion, nor a motivation, to select three non-ionic phosphorescent repeating units, in particular units as defined in claim 1, so as to arrive at a phosphorescent compound according to present claim 1. Therefore, D1 by itself does not render the subject-matter claimed obvious.

6.5.3 The only other prior art documents disclosing neutral phosphorescent compounds comprising at least one metal complex as defined in claim 1 are D4 and A2. However, none of those documents discloses polymeric phosphorescent compounds. Both documents merely disclose the use of non-polymeric phosphorescent compounds as dopants i.e. in the form of dispersed particles in a polymeric matrix. Therefore, the

combination of D1 with either D4 or A2 does not lead to the subject-matter now being claimed.

6.5.4 None of the other documents on file discloses a non-ionic polymer comprising a phosphorescent unit comprising a phosphorescent site being a complex of a transition metal or a rare earth metal as defined in claim 1. None of those documents further shows that it was obvious, at the priority date of the present application, to use a phosphorescent compound according to either D4 or A2 as a phosphorescent site in a repeating unit of a polymeric compound, let alone to use three of such units specifically emitting in blue, green and red in a single polymer.

In this regard, none of the cited documents discloses a method for the preparation of a compound containing a phosphorescent complex of a transition metal or a rare earth metal as disclosed in those documents, using such compound as a repeating unit for making a polymeric phosphorescent compound. Furthermore, Examples 6-1 to 6-4 of the present application render plausible that specific, non-obvious preparation processes have to be followed to prepare each of the monomers or the polymeric compound being claimed.

6.5.5 In view of these considerations, none of the cited documents contains a suggestion of the solution proposed by claim 1 in order to solve the above-defined problem. The subject-matter of claim 1, as well as that of dependent claims 2-11, is therefore inventive.

7. Auxiliary request 1 of the appellant (patent proprietor) being allowable there is no need for the Board to consider auxiliary request 2.

Order

For these reasons it is decided that:

1. The decision under appeal is set aside.

2. The case is remitted to the department of first instance with the order to grant a patent on the basis of Auxiliary Request 1 (claims 1-11) as filed during the oral proceedings on 15 October 2012 and to adapt the description accordingly.

The Registrar: The Chairman:

E. Görgmaier B. ter Laan