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# Datasheet for the decision of 9 November 2017

T 0473/16 - 3.3.10 Case Number:

Application Number: 02796632.4

Publication Number: 1478715

IPC: C09K11/08, C09K11/75, G06K7/12,

G07D7/12, G06K19/10

Language of the proceedings: ΕN

### Title of invention:

ANTI-STOKES FLUORESCENT MATERIAL COMPOSITION

## Patent Proprietor:

Honeywell International Inc.

## Opponent:

Bundesdruckerei GmbH

## Headword:

# Relevant legal provisions:

EPC Art. 100(a), 54(3), 56 EPC R. 99(2) EPC 1973 Art. 54(4)

## Keyword:

Grounds for opposition - lack of patentability (no)

Dec			

Catchword:



# Beschwerdekammern Boards of Appeal Chambres de recours

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Case Number: T 0473/16 - 3.3.10

DECISION
of Technical Board of Appeal 3.3.10
of 9 November 2017

Appellant: Bundesdruckerei GmbH

(Opponent) Oranienstr. 91 10969 Berlin (DE)

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Respondent: Honeywell International Inc.

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Decision under appeal: Decision of the Opposition Division of the

European Patent Office posted on 19 January 2016 rejecting the opposition filed against European patent No. 1478715 pursuant to Article 101(2)

EPC.

# Composition of the Board:

**Chairman** P. Gryczka **Members:** R. Pérez Carlón

F. Blumer

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# Summary of Facts and Submissions

- I. The appellant (opponent) lodged an appeal against the decision of the opposition division to reject the opposition against European patent No. 1 478 715.
- II. Notice of opposition had been filed on the ground of lack of novelty and inventive step (Article 100(a) EPC).
- III. The documents filed during the opposition proceedings included the following:
  - D1 EP 1 241 242 A2
  - D2 Hao, Zhao et al. Proceedings of SPIE The International society for Optical Engineering, vol. 2897, 7 November 1996.
  - D3 Zhang Long et al., Fluorescence mechanisms of  $Tm^{3+}$  and  $Yb^{3+}/Tm^{3+}$  doped AlF<sub>3</sub>-based fluoride glass, Chinese Physics, 2001, 10(01), 58-64.
  - D4 Page et al., Upconversion-pumped luminescence efficiency of rare-earth-doped hosts sensitized with trivalent ytterbium, UCRL-CR-128099 preprint, July 26, 1997
  - D6 Experimental report filed by the respondent during examination proceedings with letter dated 24 June 2010.
  - D7 Yocom et al., Rare-earth-doped oxysulfides for GaAs-pumped luminescent devices, Metallurgical Transactions, 1971, 2, 763-767
- IV. Claim 1 of the patent as granted, which is the respondent's (patent proprietor) main request in these appeal proceedings, reads as follows:

<sup>&</sup>quot;Anti-stokes fluorescent material composition which

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comprises the chemical elements M, ytterbium, thulium, oxygen and sulphur in a molar ratio relative to each other corresponding to the following formula (I), wherein M is selected from the group consisting of yttrium, gadolinium and lanthanum

$$(M_X, Yb_Y, Tm_Z)_2O_2S$$
 (I)

and wherein x + y + z = 1, x > 0, y > 0 and z is from 0,0025 [sic] to 0.03."

V. The opposition division concluded that the anti-stokes fluorescent material composition of claim 1 was novel over that of document D1, as the required amount of thulium was a selection over the general formula of D1, associated with an effect in terms of the IR/blue emission ratio.

The division further concluded that, irrespective of whether document D4 or D7 was considered the closest prior art, the problem underlying the claimed invention was to provide a fluorescent material composition having a reduced blue and an enhanced infrared emission. The solution, which was characterised by the relative amount of thulium required by claim 1, was inventive as the skilled person would not have found in any of the documents on file a reason to choose such specific amount of thulium in order to decrease the blue emission and enhance the infrared emission of an anti-stokes material.

VI. The arguments of the appellant relevant for the present decision were the following:

Paragraph [0014] of document D1 disclosed the formula  $(Gd_{1-x-y})_2O_2S:Yb_x$ ,  $Tm_y$ , wherein  $0.0001 \le y \le 0.10$ . Even

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though this formula was wrong, the skilled reader would recognise that it represented a simplification of the material's chemical formula. The sole relevant feature of such a material was the amount of dopants, which was defined by the sub-indexes x and y. As the amount of thulium required by claim 1 did not fulfil the criteria required for a selection invention, the claimed antistokes fluorescent material composition was not novel over that disclosed in D1.

Document D7 was the closest prior art. It disclosed all the features of claim 1 with the exception of the required amount of thulium. Notwithstanding that the problem of providing an anti-stokes fluorescent material having a strong IR emission and a reduced amount of blue emission was credibly solved by the composition of claim 1, such an improvement did not represent any qualitative effect for the intended use, as the blue emission of the material of D7 was already very low.

The solution, which was characterised by a defined amount of thulium, was a straightforward choice for a person skilled in the art, as documents D3 and D4 hinted at it in the context of anti-stokes materials. The claimed process was thus not inventive.

VII. The arguments of the respondent relevant for the present decision were the following:

The notice of appeal merely referred back to the arguments provided by the appellant during the opposition proceedings and did not address the contested decision. For this reason, the appeal should be considered inadmissible.

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Both general formulae in document D1 were wrong, and neither of them directly and unambiguously disclosed the claimed compositions, which were thus novel.

Document D4 was the closest prior art. However, if D7 were nevertheless to be considered closer, the problem underlying the claimed invention was to provide an anti-stokes fluorescent material composition having, like that of D7, a strong IR emission after excitation, but emitting a reduced amount of blue light. In view of the experimental data on file, this problem was credibly solved by the compositions of claim 1, characterised by requiring a specific amount of thulium. The prior art did not hint at these amounts, and thus the claimed solution was inventive.

- VIII. The board informed the parties with a communication dated 27 September 2017 *inter alia* that it tended to consider that the appeal was admissible, and that document D7 appeared to be the closest prior art.
- IX. Oral proceedings before the board of appeal took place on 9 November 2017.
- X. The final requests of the parties were the following:
  - The appellant requested that the decision under appeal be set aside and European patent
    No. 1 478 715 revoked.
  - The respondent requested that the appeal not be admitted. In the alternative, it requested that the appeal be dismissed or, subsidiarily, that the patent be maintained in the form of one of auxiliary requests 1 to 16, auxiliary requests 1 to 15 having been filed with the response to the

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grounds of appeal dated 9 September 2016, and auxiliary request 16 filed under cover of a letter dated 12 October 2017.

XI. At the end of the oral proceedings, the decision was announced.

## Reasons for the Decision

1. Admissibility of the appeal

The respondent requested that the appeal be considered inadmissible, as it was not sufficiently substantiated.

Although the appellant also referred in its grounds of appeal to its submissions during opposition proceedings, the grounds of appeal challenges different aspects of the contested decision and is not a mere repetition of the arguments filed in opposition (see for example first paragraph on page 5 of the grounds of appeal, in which the appellant contends that the opposition division did not gave any reason as to why the subject-matter of claim 1 was a narrow selection over the anti-stokes composition of D1).

For this reason, the appeal complies with Rule 99(2) EPC and is considered admissible.

- 2. Novelty
- 2.1 It is not disputed that document D1 is prior art according to Article 54(3) EPC.

Document D1 discloses an anti-stokes luminescent composition [0008] represented by two formulae which

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should refer to the same composition, namely

$$(Gd_{1-x-y}Yb_xTm_x)_2O_2S$$
, and  $(Gd_{1-x-y})_2O_2S$ :  $Yb_x$ ,  $Tm_y$ .

Only the latter formula could be relevant for the novelty of claim 1, as it is the only one for which a specific range of y values is disclosed [0014].

It is not disputed that neither of these formulae is accurate, as the sum of the amounts of Gd, Yb and Tm should be 2 for the stoichiometry to be correct.

The second formula could be understood either as  $(Gd_{1-x/2-y/2})_2O_2S:Yb_x$ ,  $Tm_y$  or as  $(Gd_{1-x-y})_2O_2S:Yb_{2x}$ ,  $Tm_{2y}$ .

- The appellant argued that the relevant feature of the composition represented by such a formula was the amount of dopants, in the present case the amount of Yb and Tm. For this reason, the skilled reader would only have considered the formula  $(Gd_{1-x/2-y/2})_2O_2S:Yb_x,Tm_y$ . This was consistent with the use, at the beginning of paragraph [0014], of the formula  $Gd_2O_2S:Yb,Tm$ , in which the combined amount of Gd, Yb and Tm was greater than 2. Both formulae merely provided simplified notations, which the skilled person would have readily understood, and which were not uncommon in the art, as shown for example in document D4, Table 1.
- 2.3 It is not disputed that  $Gd_2O_2S$ :Yb,Tm is a usual notation in the context of doped materials (D4, Table 1; D5, Figure 7; D7, page 763, second line of section "preparation of materials"). Such a notation does not indicate the amount of dopants present in the composition.

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If the amount of dopants is specified, the notation is usually of the type in claim 1, see for example D2, first page, penultimate line; D7, Table 1 or Figure 7.

The appellant has not provided any evidence that any of the two notations was usual in the art. It cannot thus be concluded that the skilled person would follow only this interpretation in the context of the second formula of document D1.

- As it is undisputed that both formulae in document D1 are wrong, and there is no evidence on file to suggest that the skilled person would only consider plausible one interpretation of the second of them, which is the only one linked to a specific amount of thulium, the board concludes that document D1 does not directly and unambiguously disclose the anti-stokes fluoroescent material composition of claim 1, with the consequence that the ground under Article 100(a) EPC in combination with Article 54(1) and (3) EPC does not preclude the maintenance of the patent as granted.
- 3. Inventive step
- 3.1 Closest prior art

The appellant considered that document D7 was the closest prior art.

It is not disputed that document D7 discloses an antistokes fluorescent composition (Figure 7) which differs from that of claim 1 by virtue of the amount of thulium that it contains (z=0.001), which is lower than required by claim 1.

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# 3.2 Technical problem underlying the invention

At the oral proceedings before the board, the respondent defined the technical problem underlying the claimed invention as being to provide an anti-stokes fluorescent material composition having, like those of the prior art, a strong IR emission after excitation, but lower blue light emission than those of the prior art.

### 3.3 Solution

The solution to this technical problem is the claimed anti-stokes fluorescent material composition, characterised in that the amount of thulium represented by z is from 0.0025 to 0.03.

# 3.4 Success

At the oral proceedings before the board, the appellant did not dispute that the results obtained in examples 1 to 3, 9, 14-15, in the comparative example of the patent in suit, and in the experimental data filed as document D6, showed that the problem formulated in point 3.2 above had been credibly solved by the compositions of claim 1, having defined amounts of thulium.

3.4.1 Despite acknowledging that the claimed compositions emitted less blue light, the appellant argued that such an effect did not represent any qualitative difference between the claimed compounds and those of the art, as the blue emission of the compound of D7 was already very low, and thus the change of the emission of the claimed compositions with respect to those of D7 was irrelevant for the intended use as a document marker.

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However, the appellant has failed to provide evidence showing that the reduction of blue light, which can be measured (examples of the application), and which the appellant has not challenged, did not have any effect on the final use of the claimed composition. For this reason, the board can only conclude that the reduction in the blue emission achieved by the claimed composition does provide an effect which needs to be taken into account when assessing inventive step.

- 3.5 It thus remains to be decided whether or not the proposed solution to the objective problem defined above is obvious from the prior art.
- 3.5.1 The appellant argued that document D7 referred to halide matrixes in the context of anti-stokes compositions. For this reason, the skilled person would have looked at the teaching of documents D3 and D4, and arrived at the claimed subject-matter without using inventive skills.

It has not been disputed that the matrix of a doped phosphor affects its properties, as is shown by comparing the results of Fig 2(c) of D4, which discloses emission spectra of different matrixes doped with ytterbium and thulium. Therefore, the skilled person would not have expected that any result achievable with a doped fluoride matrix would also be obtained using the same dopants in the matrix required by claim 1 ( $GaO_2S$ ). For this reason alone, it is concluded that the claimed compositions are inventive, within the meaning of Article 56 EPC.

3.5.2 The appellant relied on Figure 6 of document D3 and the text preceding said figure, which refers to it, in

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order to show that the claimed compositions were not inventive. The text indicates that the 476 nm emission (blue) had a maximum value at 0.1 mol% Tm, whereas the infrared emission at 797 nm had a maximum value at 0.5 mol% Tm.

Figure 6, however, shows exactly the opposite, as the lines depicted as triangles, which represent the  ${}^3F_4$  to  ${}^3H_6$  transition (corresponding to the 797 nm - infrared emission; see Figure 8), show a maximum at 0.1 mol%, whereas the  ${}^1G_4$  to  ${}^3H_6$  emission (476 nm - blue; see Figure 8) has a maximum at 5 mol%. Thus, the results provided in D3 are contradictory. The appellant has not provided any evidence, for example by repeating the experiments carried out in D3, to show which of the teachings in D3 is correct.

Thus, even if the skilled person had turned to D3, which for the reason given in the previous point the board does not consider he would have done, since it relates to a different matrix, he would not have arrived at the claimed invention.

3.5.3 Document D4 is silent about any infrared emission of the phosphors it discloses. The appellant concludes, however, from the energy levels depicted in Figure 4, that such an emission would necessarily be present. The second full paragraph on page 9 of document D4 discloses that high upconversion efficiencies required a high ratio of Yb to Tm ions, so that Tm density should be hundreds of times smaller than Yb concentration. In order to quench any blue emission, the skilled person would seek a lower upconversion and choose a lower Yb to Tm ratio, e.g. 10:1. As the amount of Yb in D7 was 0.13, the skilled person would have considered z = 0.013 and thus arrived at the claimed

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invention.

However, there is no apparent reason why the skilled person would have chosen a tenfold excess of ytterbium rather than a fivefold one, or an equimolecular ratio, or an excess of thulium. For this reason alone, the appellant's argument is not convincing.

Furthermore, there is no reason why the skilled person would have chosen any of these relative amounts in the expectation of obtaining a weaker blue emission but maintaining the IR emission. Document D4 refers to upconversion, but is silent about its effect on the relative intensity of the infrared and blue emissions. Thus, even if the skilled person had considered the teaching of D4, which for the reasons given in point 3.5.1 above the board considers he would not, as it relates to a different matrix, he would not have arrived at the claimed invention.

3.5.4 For these reasons, although the skilled person would not have expected that a result obtained for a halide matrix (D3, D4) would also be obtained for an oxysulfide one (D7), he would not have had any motivation to choose the amount of thulium of required by claim 1 in order to reduce the amount of blue light emitted by the claimed compositions.

Thus, neither document D3 nor D4 provides a hint towards the claimed compositions, which are therefore inventive, within the meaning of Article 56 EPC. For the same reasons, the subject-matter of independent claim 7 directed to a process for the preparation of such compositions, of claim 10 directed to their use for the authentication of an article, and of claim 12 directed to a marked article comprising them is also

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inventive.

3.6 The board thus concludes that the ground under Article 100(a) EPC in combination with Article 56 EPC does not preclude the maintenance of the patent as granted.

## Order

# For these reasons it is decided that:

1. The appeal is dismissed.

The Registrar:

The Chairman:



C. Rodríguez Rodríguez

P. Gryczka

Decision electronically authenticated