

# Green phosphorescent organic light-emitting devices based on wide bandgap host materials



Marc Lepeltier<sup>a</sup>, Didier Gignes<sup>b</sup>, Frédéric Dumur<sup>b,\*</sup>

<sup>a</sup> Institut Lavoisier de Versailles, UMR 8180 CNRS, Université de Versailles Saint-Quentin en Yvelines, 45 avenue des Etats-Unis, 78035 Versailles Cedex, France

<sup>b</sup> Aix-Marseille Université, CNRS, Institut de Chimie Radicalaire ICR, UMR 7273, F-13397 Marseille, France

## ARTICLE INFO

### Article history:

Received 24 October 2014

Received in revised form 3 December 2014

Accepted 8 December 2014

Available online 18 December 2014

### Keywords:

Iridium complex

Phosphorescence

Electroluminescence

OLED

Triplet state

## ABSTRACT

Two homoleptic iridium(III) complexes **Ir3F** and **Ir6F** were synthesized and tested as wide bandgap organometallic hosts for green phosphorescent OLEDs. While using Ir(ppy)<sub>3</sub> as the green triplet emitter, **Ir3F**-based devices clearly outperformed similar devices fabricated with the traditional CBP host. On the opposite, **Ir6F**-based devices showed lower performances than the reference device. As appealing feature, **Ir6F**-based devices showed almost no efficiency roll-off i.e., no reduction of the luminous efficiency with the current density contrarily to that commonly observed for phosphorescent OLEDs.

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## 1. Introduction

During the past decade, a great deal of interest has been devoted to develop highly emissive organic light-emitting devices (OLEDs) [1–5]. If the crucial role of the emitter in the overall device performances cannot be denied, for a given complex, improvement of the device architecture is also a key parameter to reach high efficiencies and luminances. To achieve high efficiencies, a careful selection of a proper host material is thus regarded as important as the choice of the materials introduced in the layers adjacent to the emissive one. To date, most of the hosts used to fabricate phosphorescent OLEDs (PhOLEDs) are organic and only few examples of organometallic host have been reported (see Fig. 1). Only 21 complexes have been tested as hosts for PhOLEDs. The first report mentioning the use of a coordination complex as host for a phosphorescent dopant was reported in 2004 [6,7]. In this study, bis(8-quinolinolato) phenolato-aluminum(III) complex **BAIq** was tested as host and hole-blocking layer for red phosphorescent OLEDs. Devices reached the remarkable luminous efficiency of 12 cd/A and the half-life of the devices was determined as being higher than 30,000 h, starting from an initial brightness of 700 cd/m<sup>2</sup>. High performances were notably assigned to the wide recombination zone enabled by the Al complex, its hole-blocking character favoring charge recombination with the emissive layer

[8], its remarkable thermal stability, and the short phosphorescence lifetime of the red dopant. More recently, salen aluminum complexes **H1** and **H2** were also proposed as hosts for red PhOLEDs [9]. While comparing performances with a reference device comprising 4,4-*N,N'*-dicarbazole-1,1'-biphenyl (CBP) as the host, a major improvement of the Al-based devices was demonstrated. Notably, **H1**-based devices exhibited a maximum luminous efficiency of 32.15 cd/A which is higher than that of CBP-based devices (21.15 cd/A). One major drawback of Al complexes is their low triplet energy levels that could not induce an efficient energy transfer to the guest if the triplet emitter was a wide bandgap complex. Use of Al complexes was thus limited to the design of red PhOLEDs. High triplet energy Al complexes were actively researched and this issue has only been addressed in 2013 and 2014 with **Al(ppyy)**<sub>3</sub> and **AlOx**. As specificity, triplet energy of these two complexes was sufficiently high to render these complexes capable to host blue (**Al(ppyy)**<sub>3</sub>) [10] and green (**AlOx**) [11] triplet emitters. Here again, organometallic hosts furnishing devices of improved characteristics. More generally, beryllium complexes possess higher triplet energy levels than Al complexes but also higher electron transporting characteristics as exemplified with electron mobilities close to 10<sup>-4</sup> cm<sup>2</sup>/Vs [12–14]. Beryllium complexes has thus been more widely studied as potential hosts for PhOLEDs. While using bis(10-hydroxybenzo[*h*]quinolinato) beryllium complex **Bebq**<sub>2</sub> as host, efficient red OLEDs were obtained, characterized by a luminance of 1000 cd/m<sup>2</sup> at only 4.5 V [15]. The current and power efficiencies of these red PhOLEDs were respectively of 9.7 cd/A and 6.9 lm/W, far from characteristics of

\* Corresponding author. Tel.: +33 4 91 28 27 48; fax: +33 4 91 28 87 58.

E-mail address: [frederic.dumur@univ-amu.fr](mailto:frederic.dumur@univ-amu.fr) (F. Dumur).

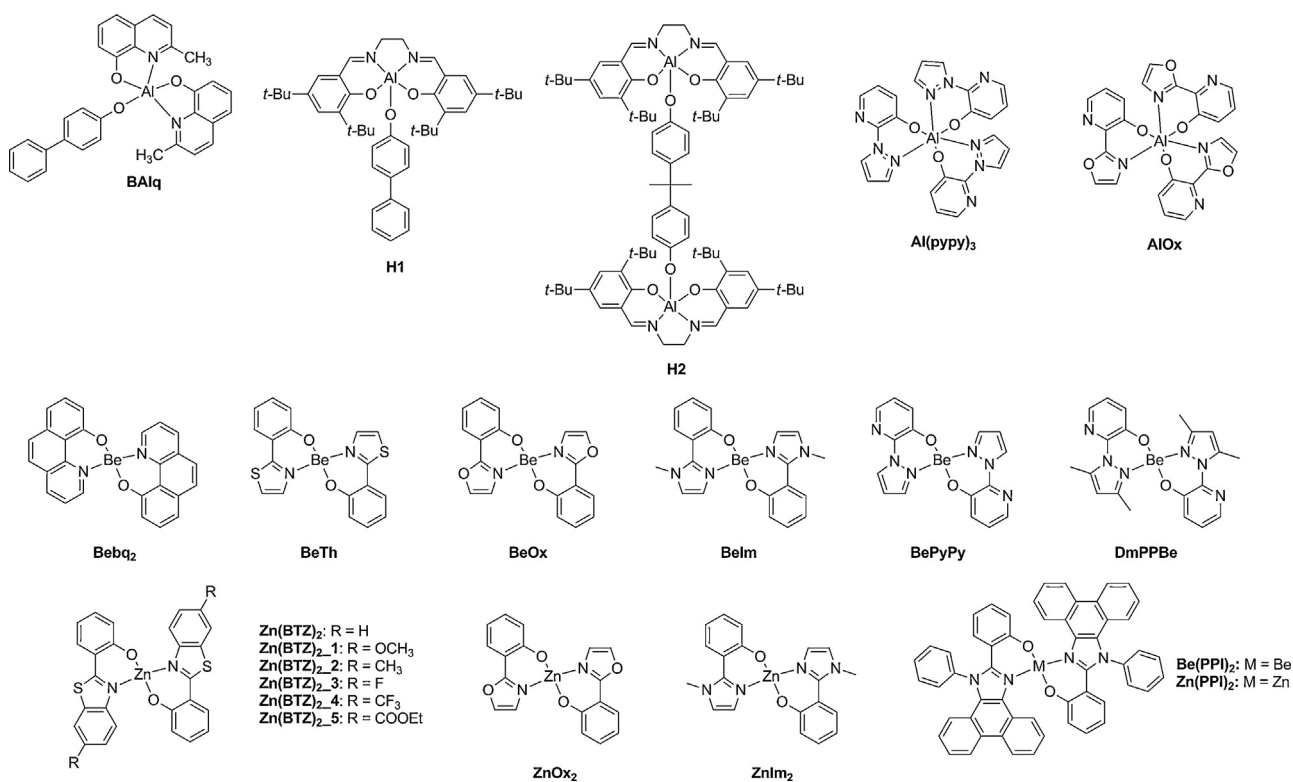


Fig. 1. Structures of the organometallic hosts previously reported in the literature.

the reference devices. In 2013, a series of triplet energy tunable organometallic hosts (**BeTh**, **BeOx** and **Belm**) was reported, enabling to prepare highly emissive PhOLEDs with emission colors ranging from red to blue (see Fig. 1) [16]. Just by simply changing of heteroatom in the ligand structure of Be complexes, triplet energy level could be modulated from 2.48 to 2.94 eV. Best results were obtained with **BeTh** comprising two 2-(thiazol-2-yl)phenolato ligand. Especially, by ligand engineering, the effective conjugation extent of the ligand could be reduced, heightening the triplet energy level. Highest triplet energy level was obtained for **Belm**. While examining the external quantum efficiencies (EQEs) of the red, green, blue and deep blue devices, values as high as 10.3%, 22.9%, 26.1% and 19.4%, were respectively determined. In the same spirit, blue phosphorescent OLEDs with an EQE of 19.7% were fabricated with the electron-transport host **BePyPy** based on pyridine-pyrazole (**pypy**) type ligand [17]. Similarly, *bis*[2-(3,5-dimethylpyrazol-1-yl)pyridin-3-olate]beryllium (**DmPPBe**) was effective as a host material for blue PhOLEDs and a high quantum efficiency of 17.7% was obtained [18]. Organometallic hosts of high triplet energy levels were also obtained with zinc complexes [19]. In this field, the first complex to be studied was *bis*(2-(2-hydroxyphenyl)benzothiazolate)zinc(II) **Zn(BTZ)<sub>2</sub>** [20]. Stable red PhOLEDs were obtained using *tris*(2-phenylisoquinoline)iridium(III) **Ir(piq)<sub>3</sub>** as the dopant. A half-luminance lifetime of 240 h at 80 mA/cm<sup>2</sup> was measured, starting from an initial luminance of 6070 cd/m<sup>2</sup>. A maximum current and power efficiency of 8.6 cd/A and 8.8 lm/W were respectively obtained, significantly higher than that measured for the equivalent CBP-based devices (2.3 cd/A and 5.2 lm/W).

Recently, the **Zn(BTZ)<sub>2</sub>** scaffold was revisited in a more systematic study with a series of Zn(II) complexes **Zn(BTZ)<sub>2\_1</sub>**–**Zn(BTZ)<sub>2\_5</sub>** varying by the substitution at the 6-position of the benzothiazole BTZ ligands [21]. Especially, influence of electron-withdrawing groups on the host ability of **Zn(BTZ)<sub>2\_1</sub>**–**Zn(BTZ)<sub>2\_5</sub>** was examined. While using *tris*(2-phenylisoquinoline) iridium(III)

**Ir(piq)<sub>3</sub>** as the red dopant, best performances were obtained with **Zn(BTZ)<sub>2\_1</sub>** and a maximum luminance of 24 870 cd/m<sup>2</sup> at 12 V, a maximum luminous and power efficiency of 13.85 cd/A and 7.25 lm/W and an EQE of 17.5% were respectively obtained, far from 9.97 cd/A, 5.22 lm/W and 12.61% obtained with CBP-based devices. Here again, devices prepared with the organometallic host clearly outperformed the organic-based devices. With reference to **BeOx** and **Belm**, the corresponding Zn-analogues **ZnOx<sub>2</sub>** and **ZnIm<sub>2</sub>** were synthesized and tested in devices [22]. A maximum power efficiency of 68.9 lm/W and a power efficiency of 57.4 lm/W at 1000 cd/m<sup>2</sup> were obtained for **ZnOx<sub>2</sub>**-based green PhOLEDs. Using **ZnIm<sub>2</sub>**, blue PhOLEDs with a maximum power efficiency of 29.6 lm/W and an EQE of 18% at 1000 cd/m<sup>2</sup> were determined. Finally, a comparative study between Be- and Zn-based organometallic hosts was reported in 2013. In this aim, two complexes were compared, namely **Be(PPI)<sub>2</sub>** and **Zn(PPI)<sub>2</sub>** (where PPI stands for 2-(1-phenyl-1*H*-phenanthro[9,10-*d*]imidazol-2-yl)phenol) that both exhibits the same HOMO and LUMO levels [23]. Several appealing features related to organometallic hosts were clearly highlighted in this study. Notably, organometallic hosts benefit from the tridimensional and rigid molecular structure inherent to coordination complexes as well as to a higher thermal stability as compared to organic hosts. The host also inherits from the intrinsic carrier-transport ability of metal complexes and the bipolar character of the organometallic host was previously mentioned in studies devoted to Al complexes. To compare the host ability of the two complexes, two iridium(III) complexes were tested as triplet emitters, i.e., a green dopant *tris*(2-phenyl-pyridine)iridium(III) (**Ir(ppy)<sub>3</sub>**) and a red dopant *bis*(2-methylidibenzof[*f,h*]quinoxaline)(acetylacetonate)iridium(III) (**Ir(MDQ)<sub>2</sub>(acac)**). Concerning green PhOLEDs, **Be(PPI)<sub>2</sub>**-based devices furnished better performances than **Zn(PPI)<sub>2</sub>**-based devices, especially for addressing the roll-off issue which corresponds to a decrease of the quantum efficiency with the current density. Conversely, similar performances were obtained with the red dopant **Ir(MDQ)<sub>2</sub>(acac)**. While

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